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L/ILW  
Management & Final Disposal

*Proceedings of*  
Sino-French Seminar, 26—28 April 1993

**BEIJING, CHINA**

# L/ILW Management & Final Disposal

Proceedings of Sino-French Seminar, 26-28 April 1993

*Sponsored by:*   BSHP, CNNC  
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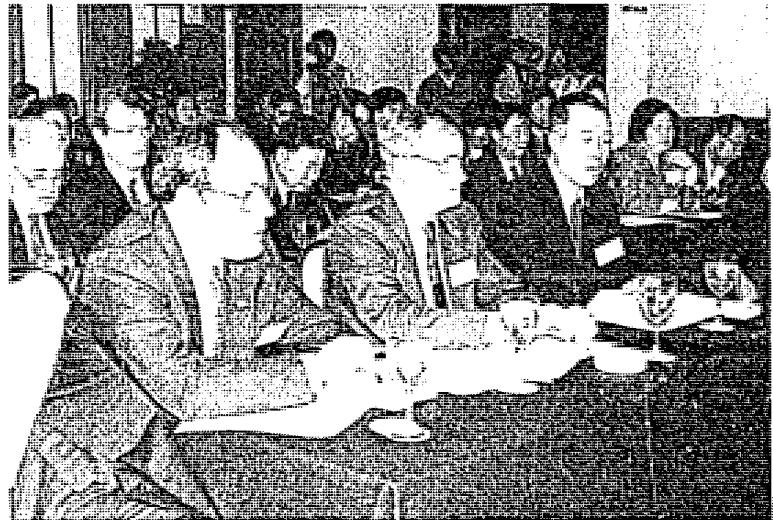
## Acknowledgements

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1. Mr. PAN Ziqiang (left), Mr. Christian DESANDRE (middle) and Mr. Yves MARQUE (right) on the rostrum
2. Mr. Thierry DUJARDIN (left), Mr. WANG Chuanying (middle) and Mr. Jean LEVIOL (right) on the rostrum
3. Participants at the Seminar
4. The rostrum

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All the participants and guests at the Sino-French L/ILW Management & Final Disposal Seminar

## Opening Statement

Dear Mr. Chairman  
Ladies and Gentlemen,

It is my pleasure to have the honour to attend this Seminar.

First, please allow me to declare that Sino-French Seminar on the Low- and Intermediate-Level Waste Management and Final Disposal is now opened.

On behalf of the Commission of Science and Technology, CNNC, I here warmly welcome our French colleagues in particular from CEA to attend this Seminar in Beijing.

As all known to us, the nuclear energy is one of the clean energy resources and plays an important role in coping with pollution from the utilization of energy. In China, the Qinshan NPP, the first NPP in mainland China, was connected to the grid and started producing electricity last year. The first unit of Daya Bay NPP is expected to be synchronized to the grid late this year. We are going to produce more nuclear energy to satisfy the growing economic development in our country. As the general public is more and more concerned about the radioactive wastes, reliable and safe management of them becomes a key issue to the nuclear energy industry.

China has already made great effort in the treatment and disposal of different level radwastes and got many achievements in the research and development works. France has rich experience in the radwaste management. The Center of Aube Disposal Repository shows the supper characterizaton.

I am fully convinced that this Seminar will benefit mutual understanding between Chinese and French experts in the field of radioactive waste management and also it will benefit further cooperation in this respect.

To end my talk, I should mention paticularly Mr. Desandre and Mr. Vienet of Technicatome and also Mr. Deng Shouchang of CNEIC for the great efforts they have made to the successful opening of the Seminar. To their valuable contribution, I would like to give my hearty thanks.

Finally, I wish great success of the Seminar.

Thank you for your attention.



Wang Chuanying

Vice Chairman

Commission of Science and Technology  
CNNC

## FOREWORD

It is a saying in France that every picture has a dark side. In the same way, every form of energy has advantages and drawbacks. Nuclear energy is no exception. In the longstanding discussions on this form of energy, management of radioactive waste has become one of the major subjects of debate and controversy in the last few years.

This issue is the major concern of the public, probably because of the accidents that occurred in recent years or were belatedly disclosed, which is a good thing, by the scientific community and the media.

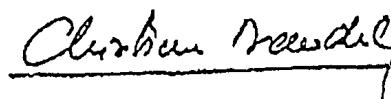
Whatever the case, this problem has solutions, not just hypothetical future ones, but real ones already implemented with success in France and the People's Republic of China.

I am pleased to state that this seminar on management of radioactive waste of medium and low activity has already allowed fruitful exchanges of information between the scientists and engineers of our two countries. I hope that the cooperation initiated will be continued in the spirit of this first seminar so as to gain increasing control over the constraints and hazards of our time and not leave them to future generations.

A number of contributions already addressed the technical solutions to the problems of processing, packaging and storing the waste, but I would also like to stress the close relationship that exists between the validity of the solutions adopted and public acceptance of them.

I would like to thank Professor Pan Ziqiang, without whom this seminar could not have taken place, and Professor Deng Shouchang and his team for their sustained effort in technical preparation and the quality of the documents produced.

Finally, I would like to thank all the participants in this seminar and hope we will soon be able to meet again in the same spirit.



Christian Desandre  
Director of Nuclear Projects  
Technicatome

## Statement

Dear Mr. Chairman  
Ladies and Gentlemen,

The Sino-French Seminar on Low- and Intermediate-Level Waste Management and Final Disposal is now opened.

On behalf of Bureau of Safety, Protection and Health of CNNC and the Chinese authorities concerned and experts in the respect of radwaste treatment and disposal present at the Seminar, I would like to warmly welcome our French counterparts to come to Beijing for the Seminar.

It is known to all that the Qinshan Nuclear Power Plant, the first one in China, was connected to the grid and started producing electricity last year; and the first unit of Daya Bay Nuclear Power Plant will be put into operation late this year. History has shown that nuclear energy is a clean energy resource and its development is an important option in dealing with energy pollution. However, because of various social and historical reasons, the general public are much concerned about the management of radioactive wastes. Reliable and safety management of the wastes has become a key issue in development of nuclear energy.

France has rich experience in radwaste management. Aube disposal repository, which started operation last year, shows the super characterization.

I am fully convinced that the exchange of technical information, through this Seminar, will benefit mutual understanding between China and France in the radioactive waste management, and naturally benefit future possible cooperation in this respect.

Here, I must mention particularly Mr. Desandre and Mr. Vienet of Tachnicatome who have made great efforts in the successful opening of the Seminar. Within such a limited time, Mr. Deng Shouchang, Manager of CNEIC Translation Service, and his colleagues, in addition to organization work, have prepared 33 papers both in English and Chinese and a dubbed video cassette of ANDRA's Aube repository. Let me show my hearty thanks to them for their valuable contribution to the Seminar.

In conclusion, I wish success of the Seminar.

Thank you.

Pan Ziqiang  
Director General  
Bureau of Safety, Protection and Health  
CNNC

Pan Ziqiang

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## **PARTICIPANTS**

**SESSION I**

**INSTITUTIONAL FRAMEWORK**

# **ESTABLISHMENT AND STATUS OF THE RADWASTE MANAGEMENT STANDARDS IN CHINA**

ZHUO Fengguan

Institute for Standardization of Nuclear Industry, CNNC

## **1. Introduction**

In the development and application of nuclear power and nuclear technologies, the safety management of radwaste is of great significance just as nuclear safety. Whether the management problem of radwaste can be solved safely and economically affects not only the health and safety of future generations, but also the prospects of nuclear power.

In the last 30 years and more, with the development of nuclear industry in China, the government, nuclear industry circles and scientists have paid great attention to the safety management of radwaste. Especially in the last 10 years, with the implementation of the nuclear power programmes, the safety management of radwaste has been legalized steadily. NEPA, NNSA, and the competent authorities concerned have engaged in establishing and promulgating policies, regulations, and a series of technical standards about the safety management of radwaste, and efforts are being made to complete and perfect related regulations and standards.

This paper introduces briefly the status and programs of radwaste safety management standards in China, including principles, organizations, and procedures for drafting the standards.

## **2. Principles for Drafting Standards**

Radwastes, especially high-level radwastes and transuranic wastes, because of their radioactive properties, will cause long-term threat or hazards to public health, safety, and environment if not properly managed and finally disposed of in safety. Hence, the management of radwaste relates not only to complicated activities in technical and economical fields, but also to social-political issues which are highly concerned by and sensitive to the general public and various social communities.

In order to ensure the safety management of radwaste, besides the general principles, the following basic principles have been formulated and followed in drafting the relevant standards.

### **2.1 Safety first**

The basic safety objective of radwaste management lies in the appropriate and optimum methodology to be used in radwaste management, so that unacceptable hazards to human health and the environment can be avoided at present as well as in the future; the design and operation of any facility and system or any activity in respect of radwaste management must meet the requirements for radiation and environmental protection, and the principles for protecting future generations.

## **2. 2 Economy**

This principle is subject to the following conditions:

- a. The feasibility in technique and economy must be considered when the specific objectives and requirements of each process in radwaste management are determined;
- b. For the standardization of a specific process in waste management, both its economic rationality and the comprehensive economic rationality in the whole waste management system shall be considered. Although there are some difficulties in this cost-benefit analysis, in any case, it is important to define this principle; and
- c. Taking into account the technical and social factors, the ALARA principle has to be implemented to keep the individual and the collective exposure lower than the specified limits.

## **2. 3 Taking disposal as the core in waste management**

The radwaste management must proceed from the requirements of the final disposal, so that the establishment of final safe disposal standards shall be, first of all, stressed, including those of the disposal technology, and the siting, design, operation, safety analysis, environmental impact assessment, monitoring, and quality assurance of disposal facility. Moreover, the standardization of other processes in waste management shall facilitate the final safety disposal of the waste.

## **2. 4 Adopting international and foreign state-of-the-arts standards according to national conditions**

It is a common principle to be followed in various projects, particularly, for the standardization of radwaste management. Firstly, it is easily accepted by all circles of the society if the international and foreign general safety principles and limits in this field are incorporated into Chinese standards. Secondly, the implementation of this principle will facilitate the international co-operation.

## **3. Organizations and Approval Procedure**

The current organizations and procedure for establishing the standards of radwaste management in China are shown in Fig. 1, which is consistent with the radwaste management system approved by the authorities concerned.

WG—Working Group

ISNI—Institute for Standardization of Nuclear Industry

CNNC—China National Nuclear Corporation

TC 58/SC 2—The National Technical Committee for Standardization of Nuclear Energy/  
Subcommittee for Radiation Protection

NEPA—National Environment Protection Agency

NNSA—National Nuclear Safety Administration

CSBTS—China State Bureau of Technical Supervision

WGD—Working Group Draft

CD—Technical Committee Draft

DGS—National Standard Draft

DEJ—Professional Standard Draft

EJ—Nuclear Professional Standard of the People's Republic of China

#### 4. Status of the Standards

A comparatively integrated and applicable system associated with the radwaste management standards has begun to take shape. It embodies the experience in radwaste management over the past 30 years in China, and reflects the generally accepted safety standards and advanced experience in international radwaste management.

At present, there are 43 standards in the system, in which 16 have been issued; 15 are to be issued; and 12 are being drafted. These standards can be classified into two categories as a whole. One includes general standards for radwaste management; and the other includes those to be used in controlling or managing the processes in radwaste management, e. g. production and discharging control, collection, treatment, volume reduction, solidification, packaging, storage, transportation, and final disposal of radwastes. Furthermore, the latter also includes some standards to be specially used in the safety management of wastes emanating from mining and milling of uranium and thorium ores.

Some typical examples of these standards are shown in Table 1.

It should be noted that most of the existing standards only serve the safety management of low- and intermediate-level radwastes. The standards for high- level radwaste disposal are being studied. This kind of standards will also be established with the issuance of the relevant policy for high-level radwaste disposal in China.

Table 1 Some Typical Radwaste Management Standards

No.	Symbol	Title	Note
1	GBxxxx	Regulation for radioactive waste management	
2	GB 9133-88	The standard for classification of radioactive waste	
3	EJ 352-88	Radioactive waste management glossary	
4	GB 13367-92	Principles for the exemption of radiation sources and practices from regulatory control	
5	GB 13695-92	Normalized limits of radioactive effluent discharges for nuclear fuel cycle facilities	
6	GB 9134-88	The technical rules for solid radioactive waste processing system of light water reactor plants	
7	GB 9135-88	The technical rules for liquid radioactive waste processing system of light water reactor plants	
8	GB 9136-88	The technical rules for gaseous radioactive waste processing system for light water reactor plants	
9	EJ xxx	The technical rules for radioactive waste treatment systems of fuel reprocessing plants	in drafting
10	EJ xxx	The technical rules for volume reduction systems of low- and intermediate-level radioactive waste	to be issued
11	GBxxxx	Characteristic requirements for solidified waste of low- and intermediate-level radioactive waste—cement solidified waste form	to be issued
12	GBxxxx	Characteristic requirements for solidified waste of low- and intermediate-level radioactive waste—plastic solidified waste form	to be issued
13	GBxxxx	Characteristic requirements for solidified waste of low- and intermediate-level radioactive waste—bitumen solidified waste form	in drafting
14	GB 7023-86	Long-term leach testing of solidified radioactive waste forms	
15	GB 13711-91	Safety Standard for low- and intermediate-level radioactive solid waste packages	
16	EJ xxx	Concrete container for low- and intermediate-level radioactive solid waste	in drafting
17	EJ xxx	Steel container for low- and intermediate-level radioactive solid waste	in drafting
18	GB 11928-89	Regulations for interim storage of low- and intermediate-level radioactive solid waste	
19	GB 11929-89	Regulations for designing storage building of high-level radioactive liquid waste	
20	GB 11806-89	Regulation for safe transport of radioactive materials	
21	GB 9132-88	Regulations for shallow-ground disposal of low- and intermediate-level radioactive solid waste	
22	GB 13600-92	Regulations for disposal of low- and intermediate-level radioactive solid waste in rock cavities	
23	GBxxxx	Site selection regulations for shallow-ground disposal of low- and intermediate-level radioactive solid waste	in drafting
24	GBxxxx	Waste acceptance criteria for low- and intermediate-level solid waste disposal facility	in drafting
25	EJ/T xxx	Standard format and content of a safety analysis report for low- and intermediate-level radioactive solid waste disposal facility	in drafting
26	GB/Txxxx	Standard format and content of environmental impact for shallow-ground disposal of low- and intermediate-level radioactive solid waste	to be issued
27	GBxxxx	Regulations for the safety management of wastes from the mining and milling of uranium and thorium ores	to be issued

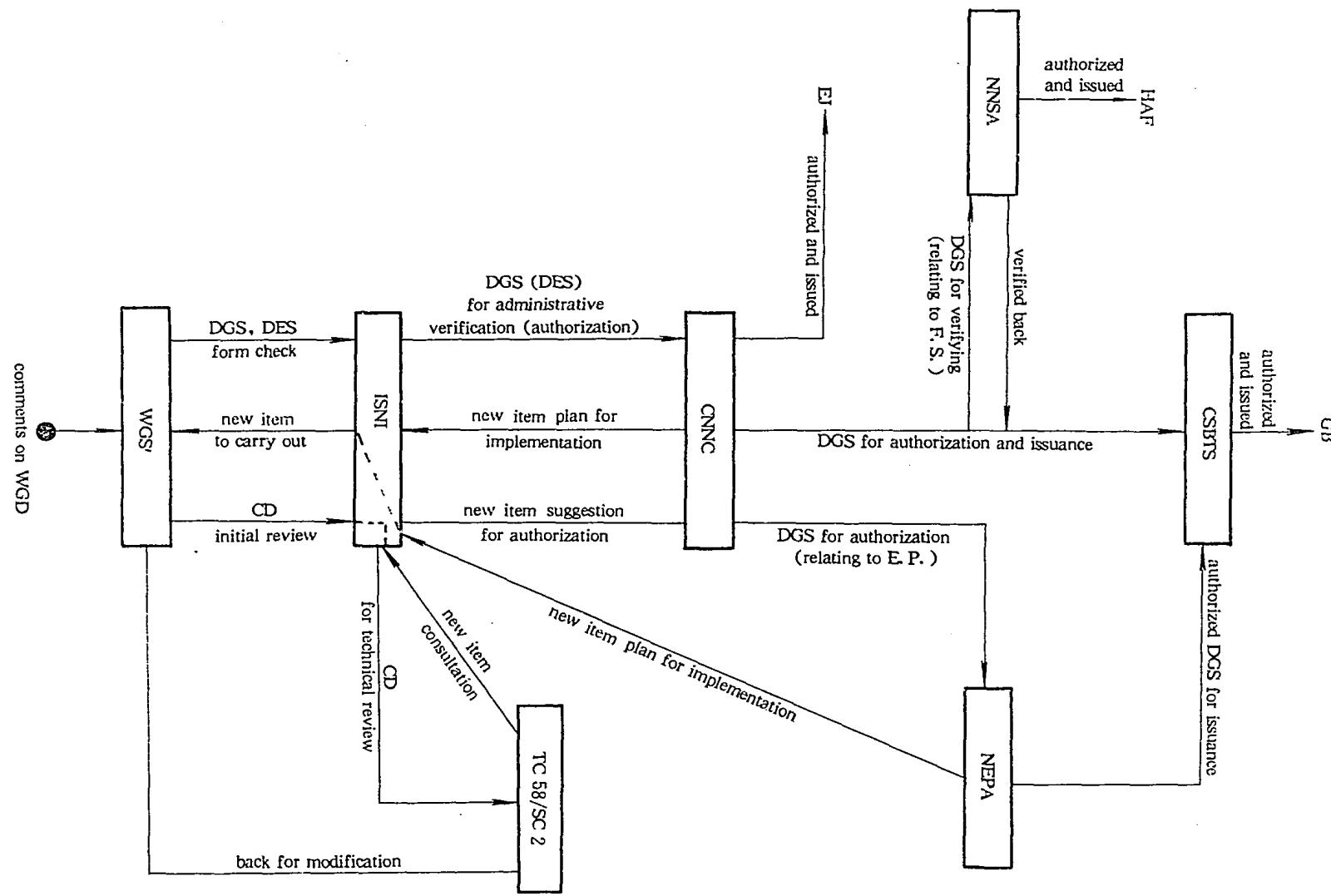


Fig. 1. The organizations and approval procedure

**SESSION II**

**ORGANIZATION IN CHARGE OF  
RADWASTE MANAGEMENT**

# **ORGANIZATION OF LOW-LEVEL WASTE MANAGEMENT WITHIN ANDRA, FRANCE**

Principles of waste management

Y. MARQUE  
ANDRA, FRANCE

## **ABSTRACT**

Short-lived waste contains relatively small quantities of radioelements with half-lives of no more than 30 years, and only trace amounts of long-lived radioelements, if any. Cobalt-60, produced by the activation of structural steel in nuclear power plants, accounts for approximately half the radioactivity in waste managed by ANDRA, yet it has only a 5-year half-life. For this reason protection from radiation emitted by this type of waste is not difficult; and the waste will become harmless in less than 300 years.

In terms of disposal safety, the guiding principle is simply to isolate the radioactive materials from the environment by disposing of only stabilized waste packages and protecting the packages from outside forces, especially water and human intrusion.

Some countries, particularly those that have elected not to sort waste into long-lived and short-lived categories, like Germany and Switzerland, plan to dispose of all waste in deep underground repositories. This approach is sometimes a matter of convenience, as is the case for countries like Sweden and Finland, which have built repositories in the Scandinavian granite shield at nuclear power plant sites. France, Spain, the United States, Great Britain, Japan, and others dispose of short-lived waste in near-surface disposal facilities. The safety of the disposal system depends on its three fundamental building blocks: the waste package, the disposal facility, and the site.

## **Introduction**

The dawning realization that human activities generate waste and that this waste has potential impacts on our environment is a trend that has had important implications for the twilight years of the 20th century. Radioactive waste in particular is a matter of considerable concern to the public, with emotional debate often clouding the public's legitimate need for information on the subject.

In France, the magnitude of the nuclear power program and the widespread use of radioelements by the medical profession, industry and the research community called for a radioactive waste management program that was national in scope. Only a public entity can be expected to exist for the time periods required for effective radioactive waste management, and is capable of reconciling short-term goals with long-term responsibility while providing technical guarantees and maintaining objectivity in the decision-making process. Accordingly, in 1979 the French Government created a new entity within the French Atomic Energy Commission—ANDRA, the National Radioactive Waste Management Agency. By law and in accordance with the Government's policy, ANDRA was given complete responsibility for radioactive waste disposal,

including the design, siting, construction, and operation of waste disposal facilities.

A major parliamentary debate was held on the program to select a long-lived waste repository, resulting in passage of legislation on December 30, 1991. The Waste Law turned ANDRA into a public service company reporting to the Ministries of Industry, of the Environment, and of Research. At the same time, the law reaffirmed ANDRA's principle functions, particularly the development of waste form specifications with which all generators must comply. The Waste Law also extended the Agency's responsibilities through the creation of the National Waste Observatory within ANDRA, whose mission is to establish and maintain a current inventory of all radioactive waste storage and disposal sites on French territory.

### Generation of Short-Lived Waste in France

Nearly 20,000 m<sup>3</sup> of immobilized waste were shipped to ANDRA's disposal facilities in 1991. Electricité de France's 58 nuclear power reactors, representing an installed capacity of 58.5 GWe, generated nearly 315 TWh of electricity and accounted for approximately one third of the total volume of waste shipped. Light water reactors generate two types of waste:

- *process waste* is waste generated by the treatment of primary coolant (such as ion exchange resins and water filters), liquid effluent (such as evaporator concentrates and ion exchange resins with very low activity levels), and gaseous effluent (such as preliminary filters, absolute filters, and iodine traps); and
- *plant waste* is waste generated primarily by maintenance operations, such as rags, paper, cardboard, vinyl sheets and bags, pieces of wood and metal, debris, gloves and maintenance clothing.

The activity of the EDF waste, expressed in gigabecquerel, is shown below:

Total alpha activity .....	6.7
Total beta activity .....	619,895
including:	
cobalt-60 .....	295,549
cesium-137 .....	25,637
strontium-90 .....	4,058
carbon-14 .....	3,497
tritium .....	353

In addition, the La Hague and Marcoule reprocessing plants shipped 7,500 m<sup>3</sup> of waste to ANDRA's disposal facilities in 1991, consisting exclusively of plant waste from operations and maintenance activities. The activity of the reprocessing plant waste, expressed in gigabecquerel, is provided below:

Total alpha activity .....	6,570
including:	
americium-241 .....	2,299
plutonium-239 .....	395
plutonium-238 .....	717
plutonium-240 .....	342

Total beta activity .....	865,490
including:	
cobalt-60 .....	2,864
cesium-137 .....	386,790
strontium-90 .....	268,111

Nearly 5,000 m<sup>3</sup> of waste from plant operations in the front end of the fuel cycle and from all of the major nuclear research centers were also shipped.

Total alpha activity .....	1,899
including:	
americium-241 .....	505
plutonium-238 .....	56
plutonium-239 .....	759
plutonium-240 .....	277
radium-226 .....	206
Total beta activity .....	141,091
including:	
cobalt-60 .....	3,641
cesium-137 .....	128,644
strontium-90 .....	4,558
carbon-14 .....	464
tritium .....	1,517

The 1,200 users of radioelements all over France — hospitals, medical research laboratories, universities, industry, etc. — generated approximately 250 m<sup>3</sup> of waste, primarily waste contaminated with a few gigabecquerel of tritium and carbon-14.

### French Policy on Short-Lived Waste Disposal

Some countries, particularly those that have elected not to sort waste into long-lived and short-lived categories, like Germany and Switzerland, plan to dispose of all waste in deep underground repositories. This approach is sometimes a matter of convenience, as is the case for country like Sweden and Finland, which have built repositories in the Scandinavian granite shield at nuclear power plant sites. France, Spain, United States, Japan, and others dispose of short-lived waste in near-surface facilities.

France has elected to dispose of short- and intermediate-lived solid radioactive waste with low- and medium- activity levels in near-surface facilities using multiple-barrier concepts in accordance with national safety regulations. Near-surface disposal methods have gradually evolved since 1969, when the first French radioactive waste disposal facility at the Centre de la Manche began operating, and have reached maturity with the design and construction of the Centre de l'Aube.

The underlying principle of near-surface radioactive waste disposal is to protect the waste from human intrusion and from exposure to water for as long as it takes for its radioactivity to decline, through the natural process of decay, to levels that are no longer harmful to the environment. The disposal facility will be monitored for the duration of the institutional control period, not to exceed the 300 years mandated by the regulatory authorities. The institutional

control period requirement is accompanied by site-specific acceptance limits for mass and total activities for both long- and short-lived emitters.

### **Operation and Closure of the Centre de La Manche**

The Centre de la Manche, located 400 km northwest of Paris on a 12-hectare site on the tip of the Cotentin peninsula near Cherbourg, is France's first near-surface disposal facility. The CEA contracted with Infratome for site operations from 1969 to 1978.

Initially, the Centre de la Manche was designed and operated as a shallow land burial site, with trenches that were covered with a plastic liner and a bitumen-impregnated membrane. After ANDRA took over site operations in 1979, the Centre de la Manche disposal concept was gradually revised to include modular disposal units made of reinforced concrete and discrete rainwater and infiltration water collection systems.

As of December 31, 1991, a total of 484,345 m<sup>3</sup> of waste had been disposed of at the Centre de la Manche. Waste shipments to the Manche are scheduled to stop in mid-1994, at which time the maximum capacity of 525,000 m<sup>3</sup> will have been reached.

Site closure operations, which began in the summer of 1991 in the northern section, will continue in 1993 on the central section, with the southern section to be completed in 1995. In addition to dismantling facilities used in site operations, closure consists primarily of revamping rainwater and infiltration water collection systems, and especially placement of the final disposal cap to protect the disposal units from rainwater for the duration of the institutional control period.

Special attention was given to the design and placement of the disposal cap. A multiple-layer design was selected whose layers are, from bottom to top, as follows:

- a level sub-grade and a series of sloped panels in the form of a roof,
- a draining layer,
- a bitumenous membrane,
- another draining layer,
- a layer of schist designed to protect the membrane from roots and burrowing animals, and
- a layer of seeded topsoil.

The total cost of closure operations is estimated at FF 435 million in 1991 French francs, or approximately \$ 87 million.

### **Design and Construction of the Centre de L'Aube**

ANDRA submitted a national radioactive waste management program to the French Government in 1984 which recommended the construction of a second disposal facility for short-lived low- and medium-level waste. The program was approved by the government, and the decision to construct the facility was announced by the Secretary of State for Energy on June 19, 1984.

By October 1984, site selection activities had begun in the Aube and Indre Departments, and later in the Vienne Department, all of which had been determined to be suitable based on site screening studies begun in 1981.

In July 1985, following submittal of a preliminary site characterization report, the government approved ANDRA's request to continue site characterization work in the Aube Department to

gather information needed to apply for a Declaration of Public Utility — similar to eminent domain — and to prepare a license application for creation of a Basic Nuclear Facility at the Pli site. ANDRA submitted both applications toward the end of July 1986, and public hearings were held on them from September 29 to November 10, 1986.

In the meantime, the Standing Committee on Radioactive Waste Disposal met on January 26 and February 4, 1987, and approved ANDRA's Preliminary Safety Analysis Report on February 11, 1987.

The Prime Minister signed a decree proclaiming the public utility of the project on July 22, 1987, which was published under docket 168 in the July 23, 1987 edition of the *Journal Officiel* of the French Republic.

During the preliminary phase of the project, a major public information campaign was launched and the conceptual design of the facility was finalized.

Land for the site was purchased in August 1987. A site clearing permit was granted by the Ministry of Agriculture on August 24, 1987, and tree felling began immediately. ANDRA also purchased a 98-hectare site in the township of La Chaise for a reforestation project to compensate for the loss of trees at the Aube site.

A construction permit for the non-nuclear portions of the Centre de l'Aube was granted on October 11, 1988, and the construction permit for the rail terminal at Brienne le Château was granted on January 9, 1989.

The Interministerial Commission on Basic Nuclear Facilities gave full approval to the project during its February 9, 1989 meeting, and the Prime Minister signed the Authorization Decree for creation of the Centre de l'Aube as a Basic Nuclear Facility on September 4, 1989, which was published under docket 207 in the September 6, 1989 edition of the *Journal Officiel* of the French Republic. The Authorization Decree allowed construction to proceed on the nuclear portions of the Centre de l'Aube. Construction at the site and at the rail terminal lasted from 1988 to 1991.

The operating license for the Centre de l'Aube was granted by the Ministries of Industry and of the Environment on December 26, 1991, and the first container of waste was delivered to the site on January 13, 1992, almost 10 years after the project was initiated.

The total cost of the project was approximately 1.3 billion 1990 francs, or about \$260 million, for a total site capacity of 1,000,000 m<sup>3</sup> and annual waste receipts of 30,000 m<sup>3</sup>. The cost breakdown is given below:

- Site selection and characterization	8%
- Project management	20%
- Construction of the Centre de l'Aube	38%
- Waste treatment facilities	13%
- Licensing	1%
- ANDRA overhead	11%
- Public information	2%
- Site access road	3%
- Rail terminal	2%
- Taxes	2%

Annual operating expenses will be approximately 200 million francs, or about \$ 40 million, and post-operating closure expenses are projected to be approximately 700 million francs, or about \$ 140 million.

The technical requirements for the Centre de l'Aube were based on 20 years of operating experience at the Centre de la Manche, and may be summarized as follows:

- waste isolation in engineered structures built above the highest level of the water table;
- long-term protection of the disposal facility against rainwater by an impermeable cap;
- collection and monitoring of infiltration water, if any, that may have contacted the waste;
- protection of the engineered structures during operations by movable buildings until a temporary cap has been placed over them;
- development of remote operating systems to reduce personnel exposure to radiation; and
- total site management consisting of separating operating units from units under construction throughout the operating period.

### **Comprehensive Waste Management System and Quality Assurance**

ANDRA worked with waste generators, regulatory authorities, and numerous subcontractors to streamline waste operations, both technically and economically, through the development of a comprehensive waste management system that covers all activities, including waste immobilization, transportation, and disposal. The ultimate purpose of the waste management system is to ensure safety at a reasonable cost while minimizing operating constraints and environmental impacts. Underlying the waste management system is a far-reaching quality assurance program that encompasses all areas of activity and that is flexible enough to apply to all waste management strategies and to adapt to special circumstances. The waste management system provides regulatory authorities with a means of verifying the compliance of site operations, and centralizes information that can be used to satisfy the public's need to know at the same time.

There are two basic components to the comprehensive waste management system:

- cost-benefit optimization of waste immobilization, transportation, and disposal operations; and
- waste tracking, from the point of generation to its final location in the disposal facility.

The first component calls on technical specifications which were established by ANDRA based on the regulations and technical requirements of French safety authorities. The waste generators must provide ANDRA with a Waste Acceptance File, similar to a Topical Report in the U. S. , for each type of waste they plan to fabricate. The file includes the following:

- a precise description of the waste and of the immobilization process, including a description of the container ;
- a description of the characterization testing program and copies of test reports;
- the quality assurance plan for waste immobilization operations; and
- the sampling and monitoring locations accessible to ANDRA in the waste immobilization process.

Waste tracking, the second component of the waste management system, is an important means of ensuring safety and offers the additional benefit of regulating transportation operations to facilitate site management. ANDRA set up a computerized network which provides a real-time link between the various waste production sites, the disposal facilities, and ANDRA's Paris headquarters. The tracking system is used to:

- identify individual waste packages;
- determine the specific characteristics of individual waste packages — type of waste, type, and concentrations of radionuclides, immobilization method, container type, etc. ;
- compare this information to waste form characteristics in the database;
- certify the compliance of the waste package to the waste acceptance file and simultaneously authorize its shipment;
- track the waste package from the generator's facility to its location in the disposal facility; and
- maintain a current inventory of total radioactivity and individual radionuclides in the disposal facility in accordance with regulations.

The radiological inventory will form the basis of the decision, to be made at the end of the operating period, on the exact duration of the institutional control period, not to exceed 300 years.

Through back-and-forth consultation with the waste generators, real progress has been made toward reducing the volumes of waste initially generated, going from 125 m<sup>3</sup>/TWh to 40 m<sup>3</sup>/TWh in the last 10 years.

### An Active Communications Program

As a public service company, ANDRA must answer to the community. The Agency's active communications program was designed to give everyone the information he or she needs to determine ANDRA's efficacy in meeting its responsibilities. Key aspects of the program are openness and adapting the message to the audience. The program also aims at providing general information on radioactive waste management as well as at explaining ANDRA's programs and its disposal facility operations.

The selection of the Aube site and facility construction were accompanied by a locally-based public information campaign that targeted elected representatives, the media and the public at large. This voluntary policy of distributing information is being continued during the operating phase, primarily through the vector of the Local Information Commission made up of elected representatives, the media, municipal, and regional government agencies and environmental protection groups from the local community.

# **STRATEGIC REVIEW ON MANAGEMENT AND DISPOSAL OF LOW- AND INTERMEDIATE-LEVEL SOLID RADWASTES**

LI Xuequn et al.

Bureau of Safety, Protection, and Health, CNNC

## **ABSTRACT**

An overview on the actual status of solid low- and intermediate-level wastes (L/ILW) management in China is described. Some of the main problems at present are analysed. The strategies on management and disposal of the wastes are discussed in light of systematology.

A large amount of solid L/ILW and distilled residual solution to be solidified have been accumulated during the past 30 years development of nuclear industry in China. These wastes, containing fission products, activated products, and uranium and transuranium elements respectively, mainly come from nuclear reactors, spent fuel reprocessing plants, and nuclear fuel fabrication plants. In the century, solid L/ILW and solidified wastes are produced mainly by nuclear industry; but in the next century, solid wastes will be steadily produced mainly from nuclear power plants.

### **1. Status in Management of Solid L/ILW**

Progress has been achieved in management of solid L/ILW from nuclear industry:

- (1) Necessary rules and regulations have been established; wastes are preliminarily managed in different categories;
- (2) Solid waste storage facilities have been constructed and used extensively in plants;
- (3) Obvious achievements have been made for recovery of uranium from solid wastes;
- (4) Greater progress has been achieved for decontamination on the surface of scrap iron and steel;
- (5) Preliminary operation experience has been accumulated in incineration of combustible wastes;
- (6) R&D programmes are being performed in treatment technologies for solid wastes, e.g. the prototype three-directional compacting and packaging machine, pyrolytic incinerator, high-temperature melting furnace and organic waste liquid incinerator and solidification facilities, and also in operation test of package system for waste drum;
- (7) Two R&D programmes are being carried out on disposal technology of L/ILW, i.e. the option demonstration of near-surface disposal of cement-solidified ILW and the review of disposal site selection and environmental impact;
- (8) A more active discussion has been conducted on the strategy study, especially on disposal. The issue of nuclear facility decommissioning has been placed on the agenda; and
- (9) In recent years, as a result of great efforts, certain progress has been made in establishment of regulations and standards for category, management, transportation, and disposal of radwastes.

Now, major problems in solid L/ILW management include:

- (1) Regulations and standards on solid waste macro-control should be further improved;
- (2) Management framework for solid wastes should be strengthened; management organizations for waste disposal have not yet been established;
- (3) An overall economy should be considered in solid waste management;
- (4) Complete data system should be established to show the quantity, properties, treatment process, and monitoring results of solid wastes;
- (5) Improvements are required in technologies for volume reduction and package of solid wastes; and
- (6) The problem about final disposal of L/ILW remains to be solved; because the old interim storage facilities for solid wastes have been used for more than 20 years, it is very difficult to retrieve and transfer these solid wastes.

In conclusion, the solid waste management has become a weak link in radwaste management in China, further efforts should be made by authorities concerned in this respect.

There are several reasons for such a weak link. From the viewpoint of history, the main reason is insufficient awareness of potential hazards caused by solid wastes. For a long time, importance has been only attached to purification of liquid and gaseous wastes while the solid waste management, neglected. In recent years, some developed countries have transferred their radwaste management from purification of liquid and gaseous wastes to disposal of solid wastes; unfortunately, we failed to follow this development in time.

In reality, the slow progress in disposal of L/ILW in China has resulted in less development of the management of solid L/ILW. Without implementation of waste disposal, management of solid L/ILW has become an aimless activity in a sense. Now, it can be seen clearly that the overall management for solid L/ILW can be promoted only when close attention is paid to disposal of L/ILW. This case is just like the fact in early years that the close attention to the controlled release and environmental protection of liquid and gaseous wastes promoted construction of purification system for liquid and gaseous wastes.

## 2. Strategic Review on L/ILW Disposal

For strategies in disposal of L/ILW, the professional group of waste management, the CNNC Science and Technology Committee, made its recommendation for "establishment of the regional disposal sites as near the producer as possible to dispose of the wastes" in the report on Recommendations for Research Plan of Radwaste Disposal (revised edition) in 1983. A regional disposal site is regarded as a large-scale L/ILW disposal site to meet the needs of nuclear industry, nuclear power plants, and nuclear applications, which will be established at several favourable locations, taking into account the factors of safety, economy, technique, and society and the conditions of geography and communications under the overall national unified planning, and adjoining to existing or planned large-scale nuclear enterprises.

There are several possible options for disposal of L/ILW, for instance, in-situ disposal, regional disposal, national centralized disposal, etc. Policy-making in this respect should proceed from the comprehensive interests of the country, rather than the local interests of an enterprise or a sector. Furthermore, any strategical society consequence, public response, and influence on future development of nuclear energy should be taken into consideration; any strategy to be made should further facilitate the development of nuclear energy.

Establishing the regional disposal sites as near the producer as possible to dispose of the wastes is one of the principles which should be followed when strategies are made. The number of permanent contamination sources can be minimized by regional disposal strategies, which can be more easily accepted by the general public than in-situ disposal strategies. In case that the regional disposal sites are selected at favourable locations, safety in environmental radiation can be further guaranteed and the project costs, lowered. In the past years, when economic rationalization of waste disposal strategies was reviewed, the costs in waste transportation were stressed unilaterally. In fact, though transportation costs can be lowered in in-situ disposal, more costs are often needed for strengthening the engineered barriers under unfavourable environmental conditions, which means that the loss outweighs the gain. Of course, in a country like China with a vast territory, compared with the national centralized disposal option, the regional disposal option as near large-scale nuclear enterprises as possible will obviously save much transportation costs and largely reduce risks during transportation.

The position of regional disposal sites should be fixed by legislation. It is forbidden that any institution manages its own L/ILW repository, or uses its interim repository as a permanent one; and it is stipulated that all the L/ILW must be concentrated and disposed of at a regional disposal site with an operation license granted by the state. The regional disposal site shall be used for both military and civilian purposes under independent management.

In the next 10 ~ 20 years, 4 regional disposal sites, located in the east, the south, the northwest, and the southwest of China, will be set up to meet the needs in development of nuclear power and nuclear industry.

### 3. Strategic Review on Management of Solid L/ILW

The objective for management of solid L/ILW is to dispose of the wastes safely and economically. All activities for management of solid wastes shall center around and be subject to this objective. In other words, construction of a regional disposal site for L/ILW is not an isolated strategy; on the contrary, it maintains close links with the management work as a whole for solid L/ILW, and even determines and restricts other policies associated with solid waste management. Some important policy issues in management of solid L/ILW must be dealt with and solved from the viewpoint of regional disposal strategy, this is our guiding ideology.

The main contents in management of solid L/ILW can be summarized as follows: controlled generation, categorized collection, volume-reduced immobilization, reliable package, in-situ interim storage, safe transportation, and regional disposal. The interrelation among them is shown in Fig. 1.

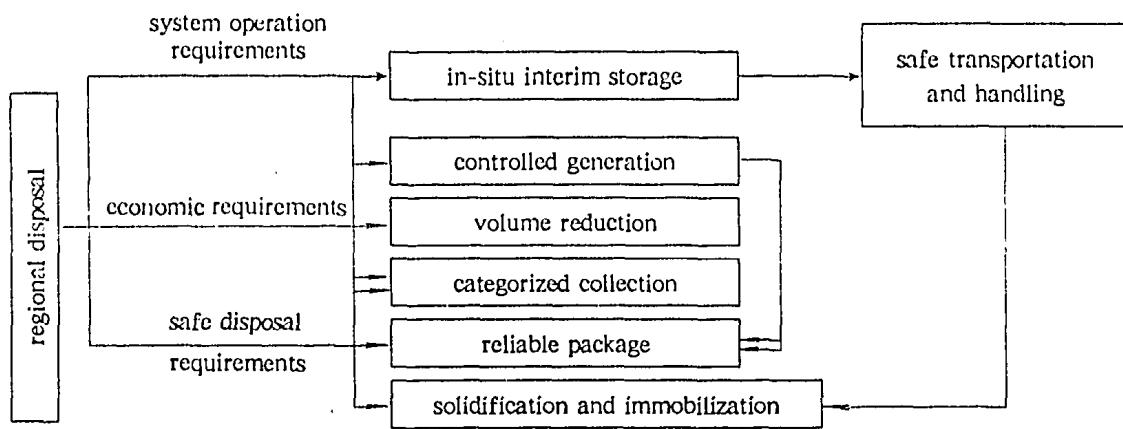


Fig. 1 Schematic Diagram of Management System for L/ILW

Recommendations are proposed in respect of management strategic issues to be solved immediately:

### 3. 1 Regional disposal

Acceptance standards of radwastes should be prepared for regional disposal sites of L/ILW, including the requirements of waste forms and package features, the refusal to accept those wastes inconsistent with the requirements, the valuation on the basis of waste categories, and the charge on the basis of waste volumes. These stipulations will greatly promote the enterprises to make every effort in control of waste generation and in waste categorization, volume reduction, decontamination, recovery, solidification, package, etc. ;

### 3. 2 In-situ interim storage

As far as the regional disposal is concerned, enterprises must set up their interim storage facilities for L/ILW, which are managed directly by themselves. In design of the facility, stress shall be paid to the convenience in retrieval of wastes, and the simplicity and practicality in structure. As stipulated in related regulations, time limit for waste transference shall be defined to guarantee that interim storage facilities will no longer become factually permanent storage facilities.

### 3. 3 Reliable package

A sound package can play various roles—safe handling, waste piling operation, possible retrieval of wastes from the storage facility, and barrier in disposing of wastes. Practice which is unable to retrieve the L/ILW, must be forbidden in case of having no package, or using only plastic bags or cartons as packaging materials. All the radwaste containers can be designed, fabricated, and used only when a license has been granted by the competent authorities.

### 3. 4 Volume reduction and immobilization

With the aim of reducing the waste volume for final disposal, controlling the waste generation and processing the wastes (compaction, cutting, incineration, decontamination, and recovery) are regarded as basic requirements in solid waste management. From the viewpoint of overall design for waste management, it is suggested that the volume-reduction technology with single-directional compaction in waste containers shall be extensively used. Despite low compaction

ratio, the technology is characterized by economy, convenience in use, and difficulty in production of the second contamination. According to acceptance standards of wastes for disposal sites in the future, not only liquid wastes but also wet solid wastes need to be solidified or dried; some dry solid forms in disperse conditions should also be immobilized, and organic liquid waste should be subjected to incineration or solidification. The volume-reduction factors are considered in selection of solidification technologies.

### **3. 5 Safe transportation**

The operation of regional disposal and interim storage system can be ensured by safe transportation; safety for transportation and handling depends on the quality of waste packages. In addition to waste packaging containers, special transportation casks must also be provided. The double-layer casks for transportation features safety in transportation as well as reduction in transportation charges. Two series of transportation casks with or without shielding shall be developed. The design, manufacture, and application of all the transportation casks can be carried out only with the related licence granted by the competent authorities. Long-distance railway transportation connected to short-distance highway transportation shall be the present approach to waste transportation. In designing and manufacturing the packaging containers and transportation casks for L/ILW, every effort should be made to ensure the transportation risks of radwastes not higher than those of dangerous chemicals, therefore the special trains and guards are no longer needed in transportation.

### **3. 6 Remedy measures**

Safety evaluation shall be conducted for the operation of existing permanent repositories and near-surface disposal sites of L/ ILW; effective remedy measures are prerequisites to meet various requirements in safety, and the monitoring and control shall be steadily.

## **SESSION III**

### **WASTE MANAGEMENT & SITING POLICIES**

# THE FRENCH LOW-LEVEL WASTE DISPOSAL SITE “CENTRE DE L'AUBE”

A ten years process, from geological concept to waste deliveries

J. C FERNIQUE  
International Relations, ANDRA, FRANCE

## ABSTRACT

The “Centre de la Manche”, first French low-level waste disposal site opened in 1969 and will enter the institutional control period around 1994.

A creation process for a new disposal was initiated in 1981 when ANDRA prepared a general radioactive waste management Program and presented it to the High Council for Nuclear Safety and Information. After acceptance of the Program, a national site screening was made, based on a conceptual geological model designed from the experience.

In 1984, the Minister of Industry announced the preselection of 3 counties out of the inventory for preliminary studies that allowed to select l'Aube as a potential site for the new French disposal “Centre de l'Aube”.

The different steps of the process and procedure that brought to the acceptance of the first packages in January 1992 will be presented in this paper, as well as the experience recently gained since that date.

This additional know-how and expertise is made available by Andra through agreements already existing with various Countries like, among others, Spain, the United States, and Mexico.

## 1. Introduction

All radioactive waste generated in France is managed by ANDRA, the National Radioactive Waste Management Agency, which was created in 1979 as part of the French Atomic Energy Commission (CEA). ANDRA's status was changed at the end of 1991 to that of a public corporation owned by the French Government.

ANDRA's principal activities are participation in research on non-land-based waste disposal; management of the existing disposal facilities; design, siting, construction, and operation of new disposal facilities; research in underground laboratories; development of waste form specifications; and preparation of a national radioactive waste inventory.

With respect to disposal, waste is normally divided into two major classes:

- the first class of waste, which is disposed of in near-surface facilities in France, includes short-lived waste containing primarily radionuclides with half-lives of less than 30 years for beta/gamma emitters and, in a few cases, a very low percentage of long-lived emitters with a mass alpha activity of less than 370 bequerels per gram; and
- the second class of waste comes from both reprocessed and unprocessed spent nuclear fuel, and may contain significant quantities of long-lived elements such as transuranics; reprocessed

fuel produces two waste types:

- alpha waste, named after its principal emitter, has low or medium activity levels and low heat releases; and
- vitrified waste, named after its solidification process, contains both long-lived emitters and substantial quantities of fission products, which release a large amount of heat that decreases over time in proportion to the approximately 30-year half-lives of cesium-137 and strontium-90.

Alpha waste and vitrified waste are stored pending final disposal. Final disposal methods for such waste are presently under study.

## 2. Near-Surface Disposal of Low-Level Waste

ANDRA developed a design concept for near-surface waste disposal in concrete structures covered with natural materials which provide complete waste isolation under controlled conditions long enough for the radioactivity to decay naturally to levels that are low enough for normal human activities to be resumed at the site. The design concept was developed beginning in 1969 at the Centre de la Manche, the first operational disposal facility in France, and was selected for the second disposal facility at the Centre de l'Aube, opened at the end of 1991.

### The Centre de la Manche

The Centre de la Manche, the first French disposal facility, has operated for 23 years on a 12-hectare site at the tip of the Cotentin peninsula near the city of Cherbourg. Depending on the radionuclide content, the type of solidification material and the type of waste container, the waste is disposed of either on concrete pads, or in reinforced concrete modules which are back-filled with grout. A final cap made of several layers of both man-made and natural impermeable materials protects the disposal structures from infiltrated rainwater. A drainage system underneath the disposal facility collects any water from the disposal units; the water can then be monitored for activity.

The principal characteristics and location of each container of waste in the disposal facility are recorded, and the record is kept in several different places for the duration of the institutional control period, which lasts several hundred years. The total amount of radioactivity contained in the waste is regularly updated. The Centre de la Manche has a total capacity of 535,000 m<sup>3</sup>; at the current waste generation rates, the site will be full in 1994 or 1995.

## 3. Centre de l'Aube Schedule

The overall schedule of the Centre de l'Aube covered a ten-year period from the end of 1981 to the end of 1991, and included the following milestones:

- Waste management program	October 1981~April 1983
- Site screening	May 1983~June 1984
- Site characterization	July 1984~July 1987
- Disposal facility construction	August 1987~December 1991.

### 3. 1 Milestone 1: comprehensive waste management program

The creation of a comprehensive radioactive waste management program can be traced to a debate in the National Assembly on October 6 and 7, 1981, which established France's energy

policy and the future direction of the nuclear power program. The waste management program has four main chapters:

- Radioactive Waste
- Waste Management Objectives
- Technical Approach
- Waste Management Program.

The waste management program was submitted to a government commission chaired by Professor Castaing in October 1982. The Castaing Commission sent the results of its review to the High Council on Nuclear Safety, which published a recommendation on April 19, 1983 calling for the search for two new near-surface disposal sites.

### **3.2 Milestone 2: site screening**

Based on the April '83 recommendation of the High Council on Nuclear Safety, on Fundamental Safety Rule 1.2 promulgated in November 1982 and published in June 1984, and on its past experience, ANDRA identified a geologic configuration type that would facilitate modelling, monitoring, and safety analysis. Gentle topographies with outcroppings of sedimentary formations in which, at the highest point, a semi-permeable formation covered an impermeable substratum were preferred. It was also preferable that the stream draining the area flow on top of the impermeable formation, collecting all site ground water, as shown in the figure.

A national inventory of potential sites was screened for the conceptual model shown above, and sites were chosen based on their seismic, geotectonic, and hydrogeologic characteristics, and on the geochemistry of the rocks.

At this point, the list of preferred sites was submitted to the Ministry of Industry for final selection and announcement to the public.

### **3.3 Milestone 3: site characterization**

On June 19, 1984, after approval of ANDRA's comprehensive waste management program, the Secretary of State for Energy publicly announced the Government's intention to construct a second disposal facility for short-lived, low-level radioactive waste. Volunteer communities to host the facility were also solicited, and the western cities of Cholet and Neuvy-le-Roi both stepped forward.

Site characterization work began in October 1984 in the Indre and Aube Departments, and later in the Department of Vienne, all of which are located in geologic regions determined to have favorable characteristics as a result of the site screening program. Site characterization began in the host community of Cholet in January 1985.

Following submittal of a precharacterization report, ANDRA was authorized to continue the Aube work in July 1985, in order to prepare a formal license application in accordance with French law. Mandatory public hearings were held from September to November 1986.

The Prime Minister signed the Declaration of Public Utility for the Aube project on July 22, 1987, thereby authorizing ANDRA to purchase land for the project. The site clearing permit was signed by the Minister of Agriculture on August 24, 1987, and the deforestation was made in the Spring of 1988. The construction permit was granted on October 11, 1988, and earth moving began immediately thereafter.

### **3.4 Milestone 4: disposal facility construction**

The design of the Centre de l'Aube disposal facility is based on the guidelines set forth in the Ministry of Industry's Fundamental Safety Rule No. 1. 2. Facility construction complies with the requirements set forth in the Decree of August 10, 1984 and its related documents. Facility operations take into account the Decree of October 2, 1986 relative to radiation protection. The design bases of the facility are the result of twenty years of operating experience at the Centre de la Manche, which may be summarized as follows:

- waste is isolated in backfilled vaults built above the highest historic level of the water table;
- the disposal facility is protected from rainwater by an impermeable cap constructed of both man-made and natural materials;
- potential infiltration water is collected and monitored;
- "suspect" water, i. e., water that may have come into contact with the waste, is eliminated by covering the disposal vault with a moveable building until it is full and a temporary cover is placed over it;
- personnel exposure is minimized by widespread use of remote operations; and
- complete control is maintained over the facility by separating vaults in operation from those under construction throughout the operating period.

When the construction permit was granted in October 1988, the site was cleared of trees and the non-nuclear facilities were built. When the site was fully licensed in September 1989, construction began on the nuclear facilities. The operating permit was granted in December 1991, and the first containers of waste were delivered to the site on January 13, 1992.

## **4. Safety Analysis**

### **4.1 Preliminary safety analysis report**

The Preliminary Safety Analysis Report (PSAR) was the first safety analysis report to be prepared when the Aube site was selected. The PSAR accompanied the license application to the regulatory authorities, submitted in 1987. The license was granted in September 1989. The PSAR consists of three volumes:

- Volume I contains general information, such as performance objectives, quality assurance organization, waste solidification, general design features and site characteristics, and environment impact assessment.
- Volume II contains a description of the site and its social and physical environment, descriptions of the waste packages and of the facilities, operating procedures, and site and environmental monitoring programs.
- Volume III contains the results of pathways analysis, based on various accident scenarios.

### **4.2 Interim safety analysis report**

The Interim Safety Analysis Report (ISAR) was prepared after the license was granted, and accompanied the application for an operating permit submitted to the regulatory authorities in January 1991. The operating permit was granted in December 1991. Like the PSAR, the ISAR consists of three volumes, but contains more details. Volume III of the ISAR, Pathways Analysis, also contains sensitivity analyses of radiological impacts in which various parameters are modified.

#### **4.3 Final safety analysis report**

The Final Safety Analysis Report (FSAR) will be prepared in the future, after the facility has operated for a few years. The FSAR will have the same table of contents as the preceding safety analysis reports, but will also contain actual results of operations and monitoring.

### **5. Waste Management System**

To ensure that safety requirements are met, while providing cost-effective disposal services to the waste generators, an integrated waste management system was developed by ANDRA which establishes a quality assurance framework for each step of waste management, from solidification to final disposal. The system is thus both a management tool to coordinate waste processing, transportation, and disposal activities, and a set of technical guidelines for acceptable operations. By integrating each step of waste management, public confidence can also be nurtured, and in fact public information is an important component of the system.

#### **5.1 Quality assurance and control**

The integrated waste tracking system requires complete control of the waste from the moment it is generated to its final disposal. A comprehensive quality assurance and control program is therefore a central feature of the system, and includes specifications for waste processing, packaging and labeling, requirements for transportation and acceptance at the disposal facility, and criteria for the siting, design, construction, and operation of the disposal facility.

ANDRA is responsible for verifying the conformance of the waste solidification process to specifications, the conformance of processing conditions to quality assurance criteria, and the conformance of the final waste form to specifications.

#### **5.2 Waste acceptance**

The first step of the system is to define waste acceptance criteria for disposal and to verify that these are met. As mentioned above, the waste has an average radionuclide half-life of less than 30 years, with limits on specific activity levels as well, and must be in stable form. In order to ensure that these criteria are satisfied by the waste generators, each processing and packaging method is thoroughly evaluated by ANDRA through a process acceptance procedure.

The waste generator must submit a "process book" to ANDRA for each waste form type. The process book includes a description of the waste stream, including the method used to determine the activity level of the waste, detailed information on the preliminary process design, the quality assurance program to be implemented, both for determining the activity of the waste stream and for waste processing, and the waste characterization program to be undertaken. The process book is reviewed by ANDRA and a group of independent technical experts during the laboratory, pilot, and full-scale characterization program before final approval is given to accept the waste processing technique.

The technical specifications relative to waste thus accepted for disposal are recorded in a "Catalogue of Waste" maintained by ANDRA as a reference manual, which includes as a minimum data on the type of the waste, the types and concentrations of radionuclides, the embedding material, the degree of immobilization achieved by the waste generator, the degree of immobilization to be performed at the disposal facility, and the type of disposal method required.

### **5.3 Inspection**

To ensure that the waste received at the disposal facility conforms to the specifications developed during the process acceptance phase, periodic inspections and quality assurance audits are performed by ANDRA. Such inspections and audits occur at the very beginning of waste generation, to verify that methods of determining the composition and activity levels of the waste stream are adequate and follow pre-established quality assurance procedures.

Inspections are also performed during waste processing, from the characterization phase through full-scale production, as well as prior to shipment and upon receipt at the disposal facility. In addition, destructive tests are carried out on each type of waste form during the characterization phase.

### **5.4 Waste generation estimating**

Long-term forecasting of the types and amounts of waste requiring disposal is needed to determine the overall disposal facility capacity requirements and its annual receiving capability. The forecasts, which are revised annually based on actual generating statistics, provide information necessary for the smooth management of disposal operations, such as the waste source, activity levels, form and packaging, as well as the type of disposal method that will be required.

### **5.5 Transportation**

Transportation, also a part of integrated waste management, must be carried out in a manner which ensures that international regulations concerning dose rates and non-fixed contamination are respected. In addition, waste transportation must be managed so that waste from dozens of locations can be delivered to one central receiving station near the disposal facility in a timely manner, while tracking each container of waste from origin to delivery and providing detailed information on its contents.

Waste is transported either by rail or by road accompanied by a shipment manifest which provides detailed information on the source, type, and activity level of each container. The manifest is checked for accuracy through physical monitoring and other means at the points of origin and delivery. Upon acceptance of the waste at the disposal facility, the data from the manifest is entered into the computerized tracking system.

### **5.6 Waste disposal**

The ultimate goal of the integrated waste management system is the safe long-term disposal of radioactive waste. By setting standards for waste forms and by verifying that these standards are met, a part of the first safety criteria relative to containment of radioactive materials has been accomplished. Now the disposal facility must complete that requirement by acting as an additional barrier against the spread of contamination. In France, this requirement led to the development of a triple-safe disposal system.

To be an efficient barrier, the disposal system must prevent water from percolating through the disposal cap and underlying structures to the waste. The cap must therefore be composed of impermeable materials, and its contours must be maintained in order to prevent subsidence and subsequent water infiltration.

As an additional measure of safety, a separative water collection network crisscrosses the vaults

to collect any water that may have infiltrated the disposal unit. The water is channeled to individual monitoring stations where it can be checked for activity and processed if necessary, thus fulfilling the "zero-release" requirement. This network provides the additional advantage of enabling any failure in the disposal cap to be detected, located, and corrected.

### 5.7 Waste tracking

Throughout each step of waste management outlined above, as well as once the waste is disposed of, thorough records must be kept on the waste, including its activity levels and form, its transportation and its final location in the disposal facility. To accomplish this, the integrated waste management system is complemented by a computerized tracking system which serves to verify the conformance of waste to specifications prior to shipment, to coordinate shipment schedules, to track waste during shipment, and to identify the location of each container of waste within the disposal system. Each waste container is labelled with a computer code which identifies its main characteristics (composition, form, activity levels, container, etc.), which is entered into the tracking system prior to shipment for comparison with the waste acceptance criteria in the data base. Once its acceptability for disposal is verified, the system can then follow the movements of that particular container of waste until it is placed in a disposal unit.

The advantage of the management tracking system is not only that it can track and locate waste, but also that it can calculate cumulative activity levels of waste disposed of at the facility, i.e. the amount of radioactivity for the facility as a whole, or for a given disposal unit, or for a category of waste, or even for a particular radionuclide.

### 5.8 Conclusions

The integrated waste management system is the result of 23 years of practical operating experience at the Centre de la Manche disposal facility. It was designed to satisfy increasingly stringent French safety requirements as well as to demonstrate to the public that radioactive waste can be safely managed over the long term. At the same time, it responds to the needs of the French nuclear program for cost-effective disposal services.

The integrated waste management system provides the framework needed to coordinate waste processing and disposal activities, while seeking and adopting ever-improved waste forms and disposal methods.

As progress is made in improving waste form, disposal structures can adjust in modular fashion to such improvements, but the fundamental principle of radioactive containment through the use of reliable barriers remains the same.

## 6. Centre de L'Aube Construction Costs

The safety of the disposal system relies to a great extent on the integrity of the waste form. A significant share of the total price paid by the waste generator goes into waste treatment and packaging, which is not reflected in the construction costs shown below. Waste transportation costs are also not included in the figures below, since the waste generator generally pays them directly. In addition, waste form and disposal technology research and development costs are not included, except for research activities relating to site selection.

## **6.1 Total construction costs**

Total construction costs for milestones 2 through 4 — site screening, site characterization, and facility construction — were approximately 1.3 billion francs (\$ 220 million) for a total facility capacity of 1 million m<sup>3</sup> of waste.

Site selection and characterization, including community incentives and public information, cost about 200 million francs (\$ 35 million). Facility construction, including the waste treatment building, the access road, and the rail terminal, cost 1.1 billion francs (\$ 185 million).

Annual operating costs will be approximately 200 million francs (\$ 35 million), including 40 million francs (\$ 7 million) for disposal vault construction.

Final closure costs are estimated at 700 million francs (\$ 120 million).

For an operating period of 30 years and a total disposal capacity of 1 million m<sup>3</sup>, the cost per cubic meter of waste breaks down as follows:

- Construction	1.3 billion francs
- Operations (30 years × 0.2)	6.0 billion francs
- Closure	0.7 billion francs
- Total for 1 million m <sup>3</sup>	8.0 billion francs

The average cost is approximately 8,000 francs per m<sup>3</sup> (\$ 38 per ft<sup>3</sup>), in 1991 currency.

The amount invoiced to the waste generator, which is based on actual costs, varies according to the form, size, and activity of the waste package.

## **6.2 Financing the construction of the centre de l'Aube**

Various options are available to finance low-level waste disposal facilities:

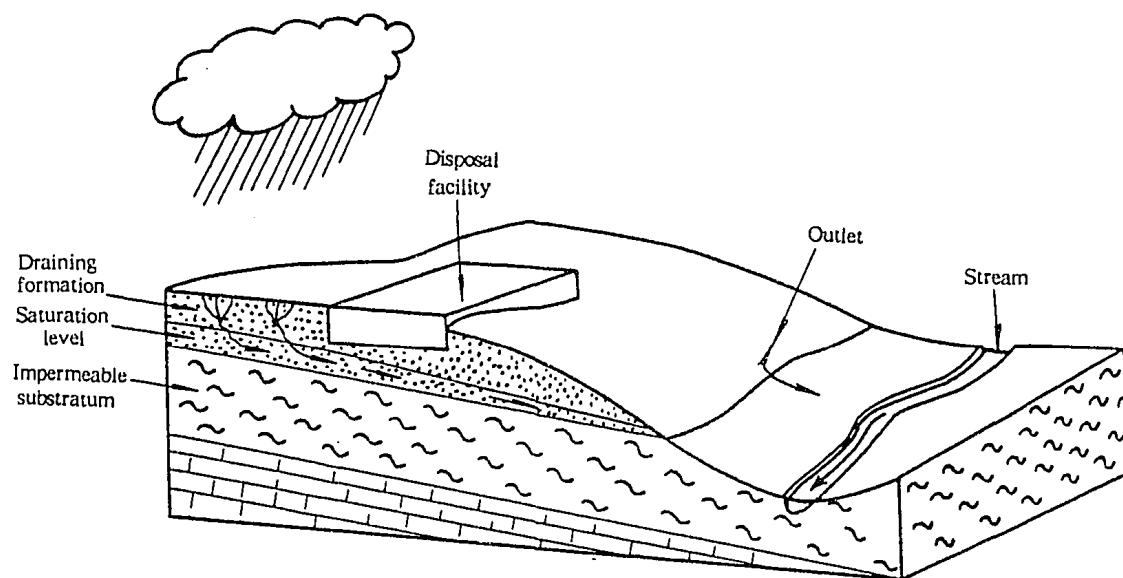
- a fee per kilowatt hour paid by the waste generator to a central organization;
- borrowing for construction and reimbursement during the operating period by a price based on units of volume; and
- direct financing of construction by the waste generators and invoicing of actual operating costs.

This last solution was the preferred one in France. A construction agreement was concluded with the principal waste generators in which the generators are essentially guaranteed a share of the facility for waste disposal, with payment based on individual waste delivery forecasts for the entire operating period. Generators are allowed to trade or sell their shares. An agreement relative to operations is also in place; an agreement relative to closure activities will be concluded in the future.

## **Conclusions**

Fifty-seven nuclear reactors are currently in operation in France. By the turn of the century, nearly 60 gigawatts of electricity will be generated by nuclear energy. The political decisions of 1981 plotted the course of waste disposal in France, resulting in the 1983 initiative to site and construct a new disposal facility for low- and medium-level, short-lived radioactive waste. Construction was completed in 1991; the final operating permit was granted at the end of that same year.

Twenty-three years of waste management experience at the Centre de la Manche disposal facility, which began operating in 1969, went into the development of a reliable and effective computer-based waste management system.



# GEOLOGICAL FACTORS OF DISPOSAL SITE SELECTION FOR LOW- AND INTERMEDIATE-LEVEL SOLID RADWASTES IN CHINA

CHEN Zhangru

Beijing Research Institute of Uranium Geology, China

With development of the modern science and technology and utilization of nuclear energy, a large amount of radioactive wastes are being generated, particularly low- and intermediate-level solid radioactive wastes generated from the nuclear power plants. Those wastes, causing harm to human health and impact on environment, have to be disposed of safely. Therefore, radioactive waste management has become one of the most pressing problems facing the world today.

As compared with other options for disposal of low- and intermediate-level solid wastes, geological disposal can be easily accepted by the public and authorities in charge of waste management. Shallow-ground disposal can provide adequate isolation of wastes from human for a fairly long period of time. The objective of disposal site selection is to ensure that the natural properties of the site together with the engineered barriers shall provide adequate isolation of radionuclides from the human beings and environment so that the whole disposal system can keep the radiological impact within an acceptable level.

Selecting a disposal site within an appropriate geological environment, regardless of the waste type or disposal option, is to a great extent based on the geological factors. In this context, there is a need to evaluate the entire geological background, comprising the lithology, stratigraphy, hydrogeology, geochemistry, geotectonics, and potential mineral resources.

In 1980s, two national standards, i.e. "Regulations for Shallow-Ground Disposal of Solid Low- and Intermediate-Level Radioactive Wastes" and "Regulations for Disposal of Solid Low- and Intermediate-Level Radioactive Wastes in Rock Cavities", were issued, and a suggestion to establish regional disposal sites for local waste has been put forward by the experts in nuclear waste management.

Since the early 1980s, complying with the national standards and the expert's conception as well as the related IAEA criteria, geological selection of disposal sites for low- and intermediate-level solid radwastes has been carried out in East China, South China, Northwest China, and Southwest China separately.

In 1970s, technology for disposal of intermediate- level liquid waste using the hydraulic fracturing was studied. The Cambrian strata which consist of sandy shale and shale have been chosen as host rocks for this kind of disposal option, where the strata are horizontal as a whole; the inclination is about  $10^{\circ}\sim 15^{\circ}$ . No faults appear in the area for hydraulic fracturing test. The main test well located at hill-side is about 500 m in depth. The findings prove the feasibility of hydraulic fracturing for disposal of intermediate-level liquid wastes in this area.

Since 1988 a program on east disposal site selection for low- and intermediate- level solid radioactive wastes has been performed by the CNNC. Based on 17 areas suitable for the disposal site, which were found in geological maps, 21 potential sites were investigated in the field.

Finally, 5 candidate sites were recommended to the CNNC.

The following criteria have been taken into account in siting:

### **1. Population Distribution**

The potential impact of radioactivity to be released from disposal site on the general public should be considered, and the distribution and characteristics of the population in the region at present and in the future should be evaluated.

Zhejiang Province is one of the densely-populated provinces in China. Its average population density is approximately 400 persons/km<sup>2</sup>. In this case, disposal site for low- and intermediate-level solid radwastes arising from Qinshan Nuclear Power Plant has to be selected in the region where population density is relatively low. In any case, cities and their surroundings, like Hangjiahu area with high population density, areas with major junctures of traffic lines, railway stations, harbours, and their neighbouring areas should be excluded from building up a disposal site.

### **2. Development of Economy and Perspectives**

As specified, the management and control period of the low- and intermediate-level wastes disposal site is generally in the range of 300 to 500 years. By then, radionuclides in the wastes can be decayed to an acceptable level before they migrate to human environment, and the land for the disposal site can be used again.

During the control period, any other activities irrelevant to waste disposal are not allowed within the site and its buffer areas.

Such areas shall be excluded from building up the disposal site for low- and intermediate-level solid radioactive waste, i.e. mineral resources areas, nature preserve zones, scenic spots, and developed economic regions.

Therefore, Tianmushan Nature Preserve Zone, Yandangshan Scenic Spot, Xin'anjiang Reservoir and its surroundings, and Hangjiahu developed economic region in Zhejiang Province are not suitable for building up the disposal site for low- and intermediate-level solid radioactive wastes.

### **3. Geological Stability**

The safety of a disposal site is highly dependent upon the engineered barriers and the geological formations which act as a natural barrier to prevent the migration of radionuclides. Favourable geological environment can minimize the contact of groundwater with the underground wastes and absorb radionuclides and retard the movement of radionuclides.

Tectonic stability constitutes a favourable condition for shallow-ground disposal sites. The area of high seismicity or in the immediate vicinity of active faults is excluded from consideration in selecting a shallow-ground disposal site.

Any active earthquake area and active fault zone as well as the area where natural disaster happens frequently shall be avoided in siting. Hangjiahu area, Zhenhai-Dinghai, Hexi-Qingyuan and Leqing-Wenzhou located in earthquake zones shall be excluded in selecting

disposal site for low- and intermediate-level solid radioactive wastes.

#### 4. Characteristics of Hydrology and Hydrogeology

The main approach of the migration of radionuclides from the disposal site into the environment is water flow, either the surface water or the groundwater. Thus, study on the characteristics of the surface water and groundwater in the disposal site and its surrounding areas aims at assessing the confining ability of the site. The potential migration approach of radionuclides and the interaction between the groundwater and rock materials can only be predicted on the basis of detailed information of the hydrogeology.

##### East China disposal site

Since the low- and intermediate-level wastes arising from Qinshan Nuclear Power Plant will be first disposed of at East China disposal site, it is suitable to select the site in Zhejiang Province. Zhejiang Province is situated in the humid subtropic zone, the rainfall is an unfavourable factor for shallow-ground disposal of low- and intermediate- level solid radioactive wastes. Hence, engineered barriers shall be strengthened to compensate for this factor.

Five candidate sites have been recommended, i.e. one shallow- ground disposal site, two abandoned zinc-lead mines, one abandoned uranium mine, and one artificial cavity at Qinshan.

The shallow-ground disposal site is preferably introduced as follows:

Bajiaotang shallow-ground disposal site is located about 200 km northwest of Qinshan Nuclear Power Plant in Anji County, Zhejiang Province .

The average population density of the township, where the site is located, is about 230 persons/km<sup>2</sup>, but the population density within or near the site is far less than that figure. Since the land is rather barren, no important crops grow there.

The topography is relatively even with an elevation of about 50~70 m above the sea level.

The lithology at the site has been found to consist of conglomerate, sandstone, and tuff with arrested occurrence. Thickness of the weathered layer is about 3~5 m. No apparent faults are discovered at the site. Earthquakes of 3~4 degree on the Richter scale are recorded. Average annual rainfall is about 1300 mm. The groundwater table is over 10 m.

In addition, bentonite and weathered tuff as backfill materials occur near the site.

##### Northwest China disposal site

In Northwest China, 6 disposal sites have been proposed in the preliminary selection, and two of them are chosen as candidate sites for characterization assessment. Both are located in stable geological units without any active faults. Lithology at the sites has been found to consist of a thick sedimentary deposit in different compositions of sand, gravels, and clay. The groundwater table is over 18 m. Average annual rainfall is below 70 mm. Population density is 10 persons/km<sup>2</sup>.

##### Southwest China disposal site

In Southwest China, 38 preliminary disposal sites have been proposed on the geological maps, 10 of them were investigated in the field; at last 3 candidate sites were selected for further investigation. They are situated in a relatively stable region, where the earthquake intensity is

below 6~7 degree on the Richter scale. The Jurassic strata, which consist of mudstone, sandstone, and siltstone, crop out at the sites.

#### **South China disposal site**

A disposal site for low- and intermediate-level solid radioactive wastes, arising from the Daya Bay Nuclear Power Plant in Guangdong Province, has been selected. 30 preliminary disposal sites were chosen on the geological maps, 20 of them were investigated in the field, 2 candidate sites, 2 km north of the Plant, have been determined. The Devonian sandy shale appears around the site. The area near Dapeng town, where the disposal site is located, is of stable geological formation suitable for construction of a disposal site for low- and intermediate-level solid radioactive wastes.

Much work and the support from the general public and the local authority are required to build up a disposal site for low- and intermediate-level solid radioactive wastes.

# CHARACTERIZATION AND HYDROGEOLOGICAL MODELLING OF A SITE FOR DISPOSAL OF MEDIUM- AND LOW-LEVEL RADIOACTIVE WASTE

J. LAVIE, P. PEAUDECERF

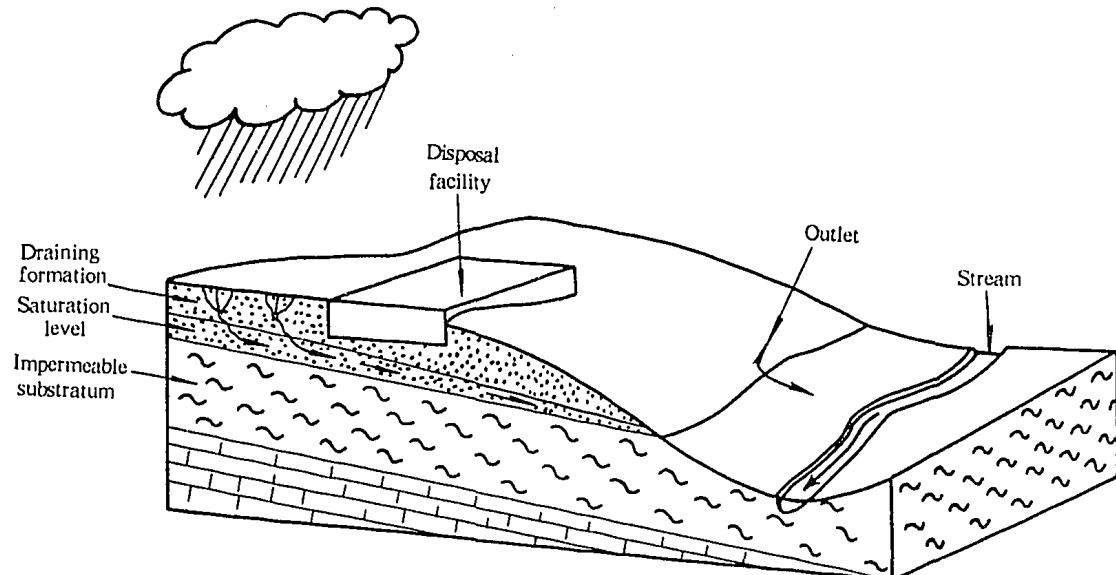
BRGM<sup>①</sup>

In order to consolidate the successive decisions that it was necessary to take concerning the various stages of authorization in the Aube Storage Centre (CSA) in France, the parameters enabling site characterization were measured, verified, and compiled using a variety of models.

The work presented herein was requested and financed by ANDRA<sup>②</sup> and undertaken by BRGM.

## 1. Measurement and Acquisition of Parameters

The hydrogeological context of the site fulfils certain prerequisites. The storage site is placed upon low-permeability clay-sand formations overlying very low permeability clays.



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① French Geological Survey

② French National Radioactive Waste Disposal Agency

The important parameters conditioning understanding of the aquifer system, its extent and its boundaries, are related to:

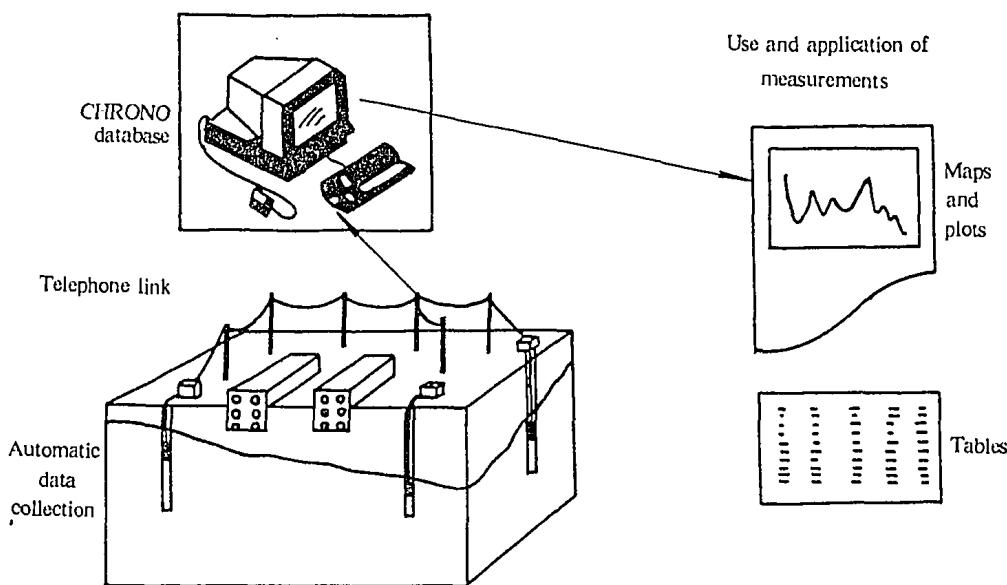
- a. piezometric levels in the various aquifers,
- b. hydrodynamic characteristics of all the formations,
- c. surface recharge, and
- d. discharges exchanged at the boundaries of the aquifer system, notably towards surface water courses.

### 1.1 Piezometric levels

Over 120 observation wells are currently being monitored within a radius of a few kilometres around the site of which 50 are on the site itself.

Measurement is either manual or automatic. The current trend is strongly towards electronic data acquisition linked to microcomputers which can be accessed directly through the telephone system (IRIS Instrument's MADO system).

Data is stored in the CHRONO-BRGM database, enabling very rapid use, access, and application including graphic output of trends, tables of measurements, and map plotting. The data base holds 90,000 measurements covering the period from 1985 to 1992 for around 300 wells.



### 1.2 Hydrodynamic characteristics

Aquifer permeabilities and storage coefficients are determined from pumping tests performed either at the outset of the study or subsequent to it in order to improve understanding as a function of project requirements.

Pumping test data are processed using the BRGM's interpretation assistance program "ISAPE" which enables a choice of type of interpretation according to the hydrogeological context using analytical solutions (Theis, Hantush Boulton, Gringarten) or a numerical, radially symmetric meshed model.

Within the framework of the present study more than 150 pumping tests have enabled definition of the hydrodynamic characteristics of the aquifer.

### 1. 3 Hydro climatological measurements

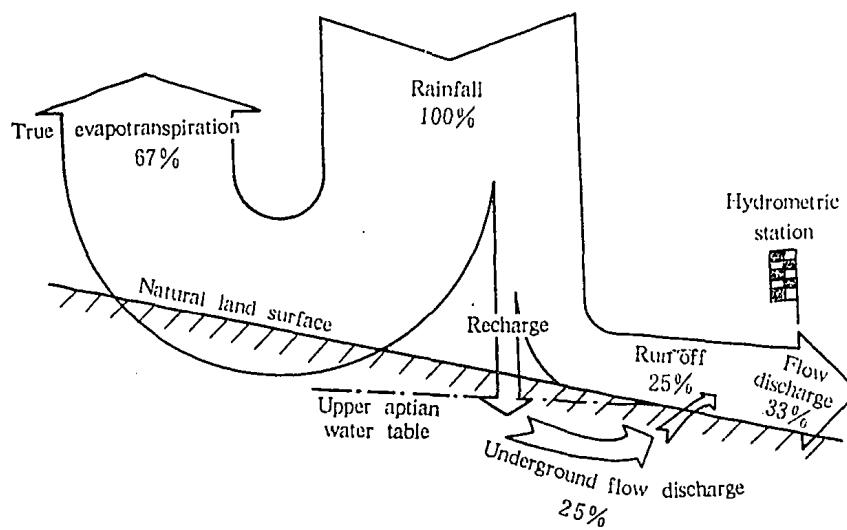
These measurements seek to determine recharge reaching the aquifer.

They concern precipitation, temperature, humidity, and insolation.

In addition, gauging of rivers is included to establish a complete hydrological balance.

The data are compiled and interpreted using a BRGM global hydrological model program called "GARDENIA" enabling calculation of hydrological parameters controlling precipitation distribution, evapotranspiration, surface water flow and recharge.

The model operates through calibration with piezometric levels and/or river flow rate.



Hydrolic balance of the storage site

## 2. Adjustment and Calibration of the Hydrodynamic Model

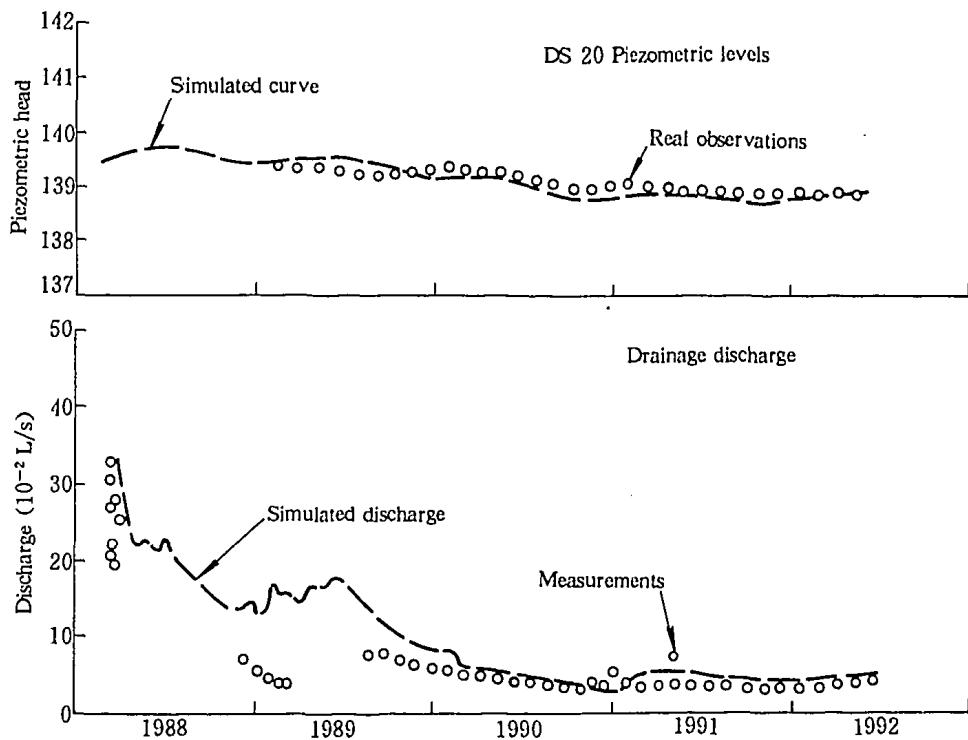
The model employed, based upon the BRGM "MARTHE" software, is a finite differences model.

The MARTHE software was developed by BRGM to simulate groundwater flow in a 2D or 3D multilayer aquifer system separated by semi-pervious aquitards. Aquifers are discretized using variable size rectangular meshes with the possibility of incorporating impervious barriers. Density variations due to salinity or temperature can be taken into account along with any unsaturated zones. MARTHE operates using the finite differences method, calculation being performed on the basis of an implicit scheme with an iterative procedure solving under steady or transient conditions.

Calibration has conventionally been by trial and error or by automatic calibration (Thiery 1993a) but recently developed options enable calibration accompanied by a sensitivity analysis of hydrodynamic parameters (Thiery 1993b).

The CSA model comprises 1,500 grid squares each 25m×25m; it is currently exploited with a time interval of 15 days.

The model has been calibrated to individual piezometric levels, piezometric maps, and drainage flow rates. The difference between model-predicted and measured groundwater levels is less than 50 cm for more than 85% of the model's surface area.



The model was calibrated for the period 1988 to 1991 and is validated every year. Model-predicted compared to measured data on recharge, piezometric levels, and flow rates are acquired for subsequent periods without changing the parameters.

The hydrodynamic model of assured reliability can thus be employed as the fundamental tool in the hydrogeological characterization of the site.

### 3. Application and Use of the Hydrodynamic Model

The hydrodynamic model, which compiles the measurements made on the site, is applied according to three main orientations:

- short term forecasting,
- medium to long-term forecasting, and
- aquifer surveillance.

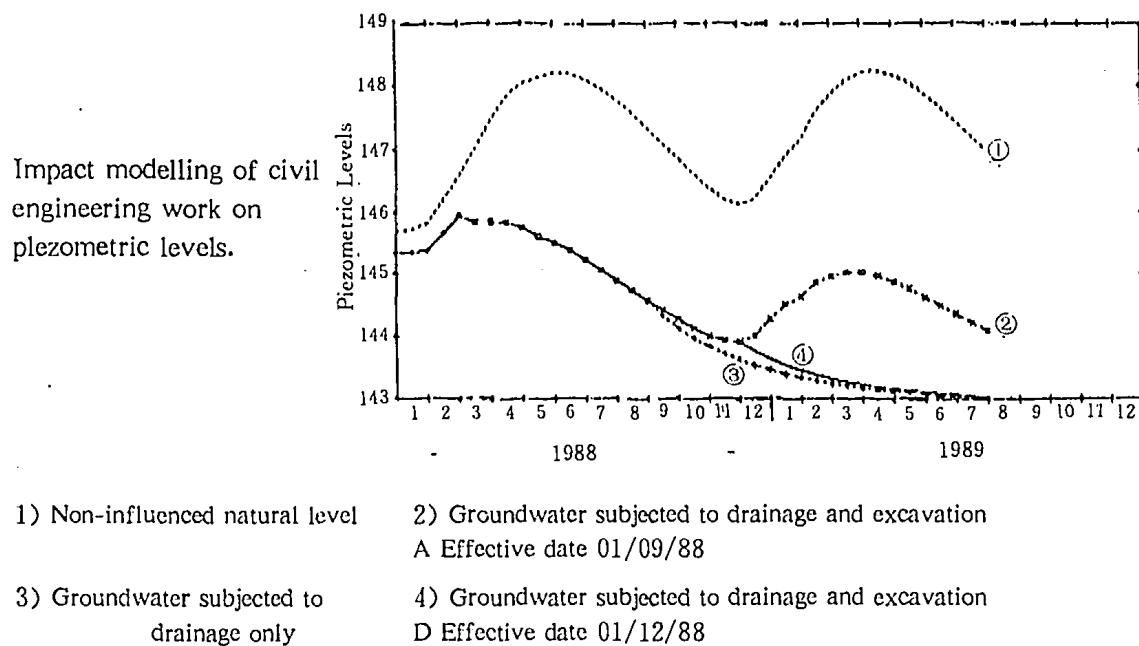
### 3.1 Short-term forecasting

This stage covers, for the most part, the Centre's operational phase. Civil engineering works planned by ANDRA are simulated using the model and a provisional evaluation of the hydrological impact determined, enabling an appraisal of the efficiency of an installation, the impact of a development and even definition of the planning phase of certain works.

As an example, an impact study for peripheral drainage installations, the purpose of which was to increase the piezometric drawdown, enabled definition of the impact and dimensioning of the size of the installation.

In addition, modelling of the impact of earthwork operations can be cited. This work stripped land naturally covered by a layer of clay and which, it was feared, and according to initial planning, was likely to cause a rise of the water table level leading to difficulties with other work on the site.

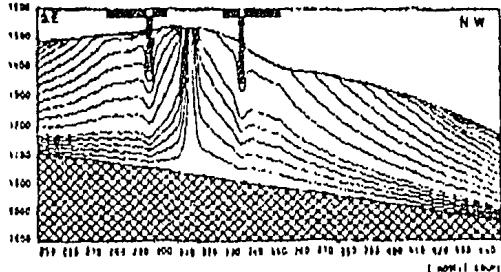
N. B. Results of the short-term predictions were verified shortly afterwards since the actual work followed modelling and enabled confirmation of the model's validity.



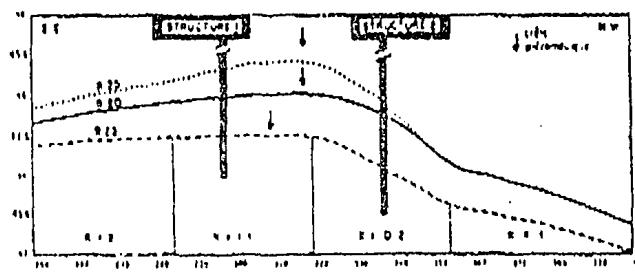
### 3.2 Medium to long-term forecasting

In this case, the hydrodynamic model is used to predict, for example, the highest water table levels resulting from exceptional recharge with a recurrence frequency of 100, 300 and 1,000 years. The results enable definition of storage levels which, according to fundamental rules of safety, should remain above the highest water levels. This type of study is based on climatological analysis and an application of the hydrological model for evaluation of recharge. In the CSA case, the study was based on analysis of precipitation statistics covering a 113-year period.

Also worthy of mention are studies undertaken to evaluate the impact of particular types of underground development on groundwater flow characteristics or the effect on the groundwater recharge regime of surface construction of impervious disposal cells.



Trajectory traces beneath storage cells



Influence on the free surface of modification to the infiltration between two structures

### 3. 3 Aquifer surveillance

The longer and the more contrasted the observation period is, the more reliable the calibration of a model will be. With this in mind and considering piezometric, hydroclimatic and discharge measurements, and data collection from the various works at the Centre is continuous. Additional data collection sites may also be installed to improve understanding of certain, insufficiently investigated zones, discovered during the modelling process. The information gathered is then incorporated into the reference model, further enhancing reliability.

## 4. Modelling of Transport

### 4. 1 Computation code

The computation code used (SESAME - BRGM) is a three dimensional solute transport model which solves the advection dispersion equation using a particle tracking method. A number of particles, each carrying a given mass of solute, undergo three successive displacements at each time interval.

- for advection and longitudinal dispersion the particles are tracked along pathlines
- for transversal dispersion (in both y and z directions) the particles are moved in a plane perpendicular to the local velocity.

In each mesh of the model, an analytical solution is used to compute the pathlines after linear interpretation of the velocity.

This code has been designed to use heads (or velocities) directly from the MARTHE software which ensures the consistency of results.

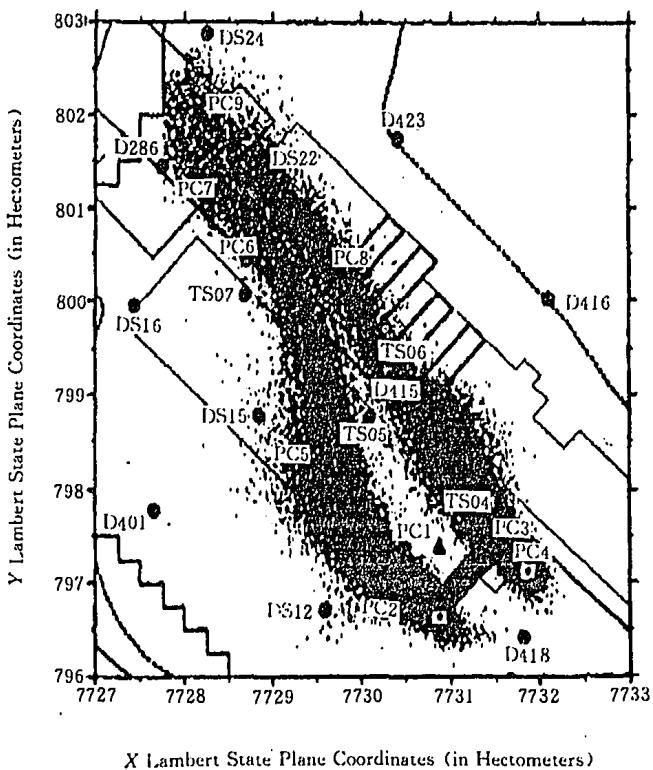
### 4. 2 Application and use of the transport model

The transport model is also an essential tool in site characterization used for forecasting studies on the migration of elements in solution and for sizing surveillance apparatus as a function of the hydrological regime of the aquifer and the configuration of the storage facility.

Experimental tracing of natural flow at site scale have enabled the hydrodispersive characteristics of the groundwater to be determined, thus improving the reliability of both the hydrodynamic and the transport models in ensuring an overall coherence of the data which describe the

functioning of the aquifer.

Plot of particles resulting from continuous (50 years) injection simulation



## Conclusions

Characterization studies of the French low- and medium-level radioactive waste site in the Aube Department includes a significant hydrogeological appraisal element. These studies are based upon geological, hydrogeological, and hydrodynamic measurements which are compiled into a model of the ground water body.

This essential tool has been used and applied throughout the lifetime of the Centre from the initial green-field site description phase to the forecasting of highest water levels applicable to both current and post utilization phases.

Data collection is continuous and the model is regularly validated providing ANDRA with a reliable and up-dated tool.

It should be noted that these results were obtained through team work based on close relations between ANDRA and BRGM enabling immediate and rapid adaptation to problems as they arose during the course of the study and as the requirements of the civil engineering works progressed.

In addition, it should be stressed that one of the essential points of interest with regard to hydrodynamic modelling is a study of the sensitivity of results within a range of realistic hypotheses and the opportunity for subsequent reorientation of operations. The ability to be in direct link with civil engineering work in progress, with real-time feed-back, is an additional and original aspect of hydrodynamic modelling.

## References

- DEWIERE (L.), LAVIE (J.), ANDRE (P.), ANDRE-JEHAN (R.), PEAUDECERF (P.), The different parts of the hydrodynamic modeling in the performance assessment of a low level radwaste repository. Safewaste 93, International Conference, 14 ~ 18 June 1993, Avignon-France.
- ANDRE-JEHAN (R.), MOLINAS (E.), Restoration of the environment and management of hazardous waste sites, 29th congrès géologique international, 24 August~3 September 1992, Kyoto (Géochronique No. 42).
- THIERY D. - 1993a - Calibration of groundwater models by optimisation of parameters in Homogeneous Geological Zones : Internat. Conf. on "Stochastic and Statistical Methods in Hydrology and Environmental Engineering", Univ. of Waterloo, Ontario.
- THIERY D - 1993b - Automatic calibration of groundwater models by the head gradient method. GQM 93. Internat. Conf. on Groundwater Quality Management. Tallinn, Estonie, Sept. 93.

# THE DISPOSAL OF INTERMEDIATE-LEVEL RADIOACTIVE LIQUID WASTE BY HYDRAULIC FRACTURING PROCESS

CHEN Ruilin ZHOU Hanchen GAO Yuzhu  
QIAO Wen WANG Wentao

Beijing Institute of Nuclear Engineering, China

## 1. Introduction

It is a preferable method that intermediate-level liquid waste (ILLW) is disposed of by the hydraulic fracturing process with low cost and large capacity of disposal, a continuous injection of 8 hours can dispose of ILLW about  $300\sim350\text{ m}^3$ , more than  $10,000\text{ m}^3$  radioactive liquid waste can be disposed of in each well, therefore it is an attractive process for the disposal of ILLW.

Since Sichuan Nuclear Fuel Plant has better geological conditions, it is possible to dispose of its ILLW by using hydraulic fracturing process. Since 1980s, the research and development of the hydraulic fracturing process have been initiated for disposal of ILLW. It is demonstrated that the hydraulic fracturing process is considered suitable for disposal of ILLW in that region.

## 2. Description of Hydraulic Fracturing Process

The hydraulic fracturing process is characterized by combination of the treatment with the disposal of ILLW. It is similar to the cement-solidified process in respect of treatment technology, but is of cement solidification in deep geology stratum. In carrying out the hydraulic fracturing process, first of all, it is necessary to select a suitable disposal site with detailed information on geology and hydrogeology. The rock stratum between the lower limit of groundwater and above 1,000 m in depth of underground will be selected as the disposal layer. The ILLW which is mixed with cement and other additives to form grout is injected into the underground closed stratum with extremely low permeability to solidify with rock to become an integral body by using matured fracturing technology and equipment available to oil industry and the technology and facilities related to nuclear safety. As a result, the purpose that the radioactive waste is isolated from human environment can be reached.

### 2. 1 Advantages of hydraulic fracturing process

Compared with the near surface disposal of ILLW, the hydraulic fracturing process has such advantages as small in building area and simple, safe, and reliable in technology. Since the treatment and the disposal of ILLW are carried out simultaneously, the cost of final disposal is much less than the others. It is clear that this process benefits the environment and mankind.

### 2. 2 Basic requirements

#### 2. 2. 1 Requirements for radioactive waste grout

- a. Liquid waste must have good chemical compatibility with cement and additives;
- b. The viscosity of the grout does not exceed  $4\times10^{-2}\text{ Pa}\cdot\text{S}$ ;
- c. After solidification, the product contains a great part of water, and the free water will not

- be allowed to exceed 3 to 5 %;
- d. The densifying time of the grout should be corresponding to the pumping time and a time of 10 to 12 hours should be guaranteed;
  - e. The setting time of the grout is not allowed to exceed 7 days;
  - f. The leaching rate of the solidified product should be very low; and
  - g. The compressive strength of the solidified product should not be less than 700 kPa.

## 2. 2. 2 Geological requirements for disposal site

Types of rock, stability of geological structure, history of earthquake, distribution of crustal stress, effective porosity of rock stratum, compressive strength of rock stratum, mineral composition in rock including their absorbability to ions and their exchange capacity, etc. must satisfy the requirements for disposing of ILLW when the hydraulic fracturing process is used.

## 2. 2. 3 Radiation protection

The rock stratum of the site to be selected should be thick enough to provide a shielding layer against radiation.

## 2. 2. 4 Maximum conceivable accident

The injecting process should be safe and reliable, and the emergency measures are provided for the maximum conceivable accident.

## 2. 3 Site selection

On the basis of the principles mentioned above, a stretch of steady mass of shale rock with complete geological construction has been discovered near the Sichuan Nuclear Fuel Plant. The shale is wide in distributive scope and large in thickness, the content of clay mineral is high, the lower limit of groundwater moving range is 100 m deep. The earthquake intensity of the area is low and the crustal stress in three-dimensional space is advantageous to the hydraulic fracturing injection.

# 3. Hydraulic Fracturing Tests with Water and Grout Injection

The water and grout injection tests started in May 1980, and about 270 m<sup>3</sup> of water and 291 m<sup>3</sup> of grout were injected. The clefts approximate to horizontal were produced in both injections.

Measured parameters are shown as follow:

Breakdown pressure: 26 MPa; Corresponding injection rate: 0.13 m<sup>3</sup>/min;

Prolongation pressure: 20 MPa; Corresponding injection rate: 1 to 1.13 m<sup>3</sup>/min.

The results of the grout injection test conformed to those of the water injection test, and the expected purpose of the test was achieved.

# 4. Concept Design for Disposal ILLW with the Hydraulic Fracturing Process

## 4. 1 Flow diagram

The flow diagram of the hydraulic fracturing process is shown in Fig. 1. Solid materials such as cement, fly ash, activated clay, and zeolite are weighed according to the prescription that is determined by the test, then they are blent and transferred into a high-level blended material tank. When the ILLW, added with retarder and deaeration agent, is pumped into a jet located at

the bottom of the mixer under a certain pressure and a certain flowrate, thanks to the action of negative pressure, the solids fall into the bottom of mixer and is mixed with ILLW in a certain solid-liquid ratio to form the grout that meets the requirements. This grout is pressured by the injection pump and flows through the coiled tubing into the injection well.

The radioactive grout with high enough pressure cleaves the shale stratum, and is solidified under pressure, thus an integral body with the rock stratum formed.

#### 4. 2 Safety measures

##### 4. 2. 1 Monitoring for operation

During each injection operation, the change of injection pressure and the exposure dosage of the operational crew must be monitored. The injection pressure is steady in general in the whole injection process, if a large fluctuation or a pressure drop which is not explainable appears, which means that a vertical cleft is probably produced, the operation shall be stopped at once and causes must be analyzed.

##### 4. 2. 2 Determination of orientation and distribution range of underground grout sheet

Orientation and distribution range of the radioactive grout in underground are determined by the micro-earthquake method, and the distribution range diagrams can be drawn out by a drafter.

##### 4. 2. 3 Observation wells for covering rock stratum

Observation wells for covering rock stratum are built around the injection well. In general, after every four injections, it is needed to observe the leakage rate of water of the naked part at the bottom of wells under 0.5 MPa pressure. If the leakage rate of water increases evidently after injecting, which means the airtightness of the rock stratum where the grout is injected is damaged, the injection shall be stopped.

##### 4. 2. 4. Determination of rise value of earth surface

In order to protect the safety of buildings and structures on the earth surface, and to know the orientation of underground grout and the change of rock stratum indirectly, the rising value of earth surface must be determined after each injection.

##### 4. 2. 5 Emergency waste pool

In order to prevent the grout which has been injected into the rock of underground from coming back to the earth surface in case that the injection pump or the high pressure manifold or the well-head device ruptures, in addition to the blowout preventer assembly, an emergency waste pool of 150 m<sup>3</sup> is established to store the radioactive grout coming back to the earth surface.

#### 5. Workshop Arrangement

The layout of main equipment of the hydraulic fracturing workshop is shown in Fig. 2.

The workshop is arranged according to the principle of four zones. The equipment for storing and handling radioactive materials are arranged in Waste Liquid Storage Room, Mixing Room, Injection Pump Room, and Well-head Device Room respectively. In addition to the concrete shielding layer which is safety enough, all rooms are lined with stainless steel cladding. Viewing windows are installed in the walls of the rooms to monitor the whole injection process and the equipment's running conditions directly.

## **6. Maintenance and Decommissioning of Equipment**

### **6. 1 Maintenance**

According to the radioactive levels of the materials to be handled, the workshop's equipment are maintained with direct and indirect methods respectively. But the method of direct maintenance is adopted for mixing equipment, injection pump, and coiled tubing, and so on which are introduced from the oil industry. Before maintained, they must be cleaned with high pressure water.

### **6. 2 Decommissioning**

When the task of hydraulic fracturing is completed, the well field shall be sealed with cement, covered by clay, cultivated with plants and marked with "Disposal Area". and it can be isolated safely, so the environment can be guaranteed not to be polluted.

## **Conclusions**

There is a stretch of mass of shale rock near Sichuan Nuclear Fuel Plant which geological structure fits the disposal of ILLW by the hydraulic fracturing process. The geology of the site is steady. The rock stratum is thick and distributes extensively, and contains rich mineral clay, so it is an ideal site.

The concept design of the process, equipment, and workshop building has clearly explained that this project is feasible, and the operation can be accomplished in safety. And the purpose of economic disposing of ILLW is achieved.

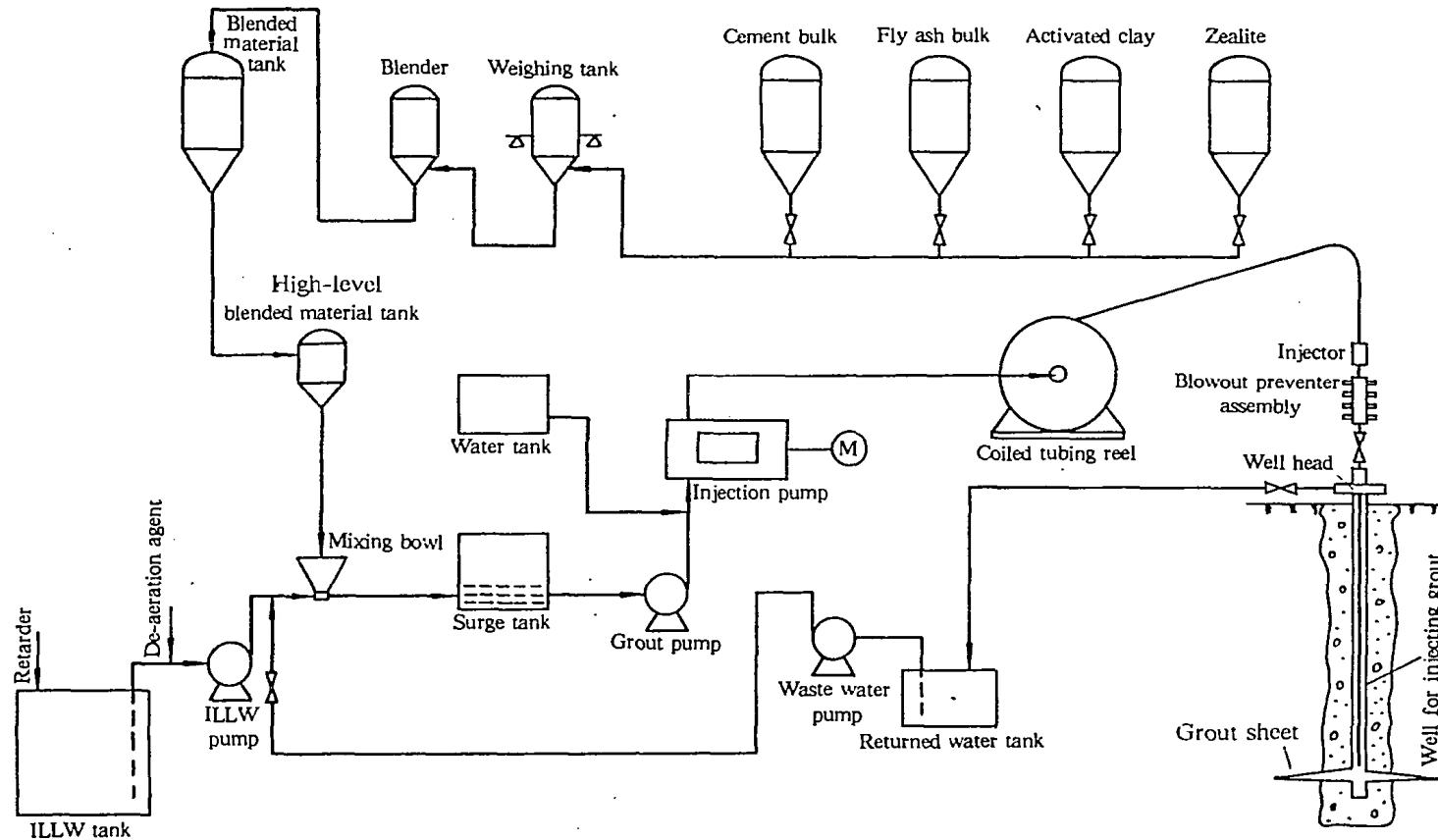


Fig. 1 Flow diagram of the hydraulic fracturing process for disposing of ILLW

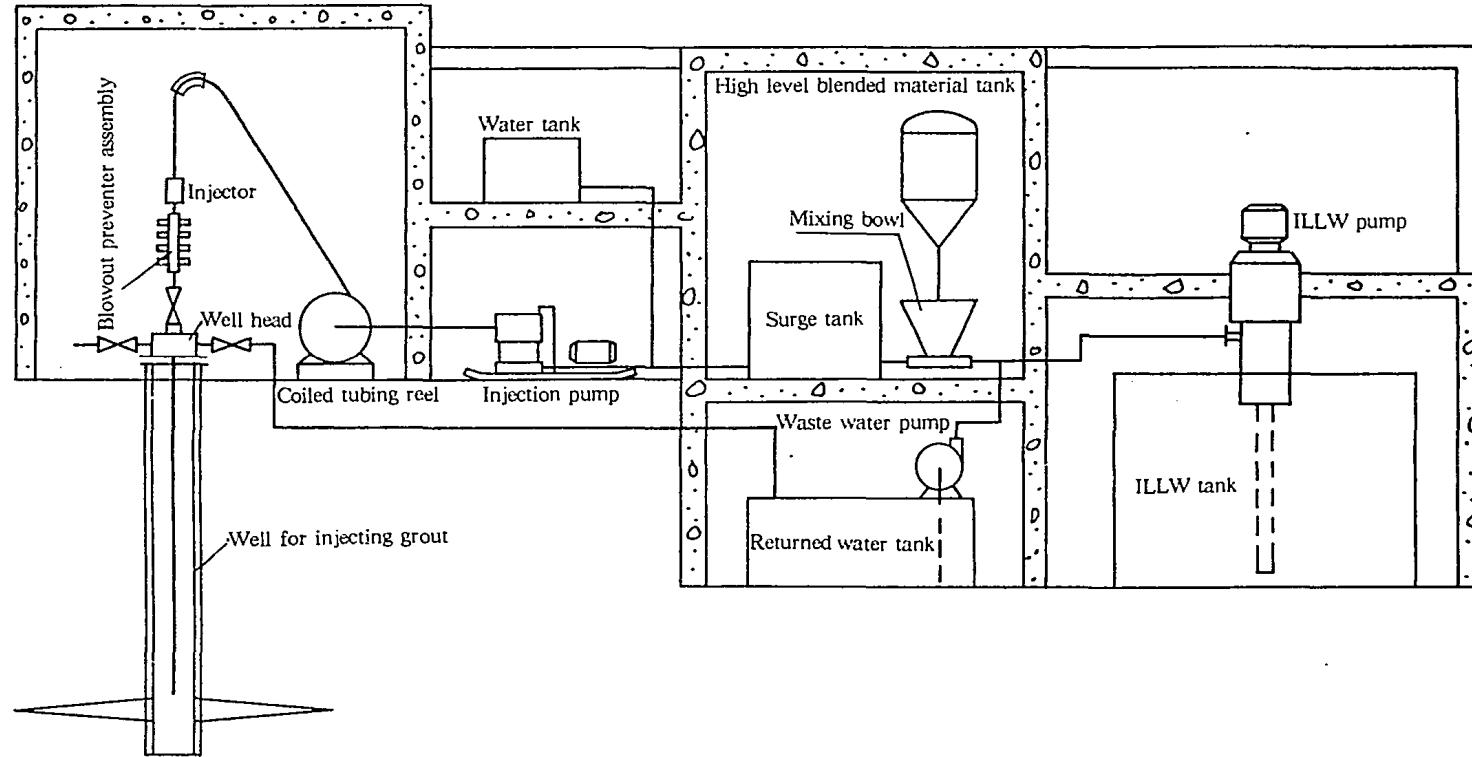


Fig. 2 Layout of workshop building for hydraulic fracturing process

# **PRELIMINARY SITE INVESTIGATION FOR LL AND IL RADWASTE DISPOSAL FOR QINSHAN NPP**

HUANG Yawen CHEN Zhangru et al.

China Institute for Radiation Protection

## **ABSTRACT**

With the purpose of selecting a disposal site for the low- and intermediate-level radwastes arising from Qinshan NPP, site investigations were carried out in several districts of Zhejiang Province. Investigation objectives included the circumstances of geology, hydrogeology, environmental ecology, and social economy. On the basis of collected data, five possible sites were recommended for policy-making reference and further investigation.

### **1. Introduction**

Qinshan NPP is the first one which is designed and constructed on domestic efforts. It is necessary to build a disposal site for the low- and intermediate-level radioactive wastes to be produced in the plant. Upon review and evaluation, it was concluded that the disposal site should be located in Zhejiang Province. Therefore, a site review group has been formed to perform a preliminary survey of the possible sites. Some references and data have been collected.

### **2. The Criteria of Siting**

#### **2. 1 Geological stabilization**

The site must be kept away from the area with serious detrimental geological conditions to waste disposal, such as faults, folds, earthquake, active volcano, collapse, erosion, subsidence, landslide, the highly-concentrated ground stress, and the earth surface rising or subsiding fast. In addition, it must be kept away from the tsunami and surge zones.

#### **2. 2 Geological formation and lithology**

The geological formation of the disposal site and its adjacent area shall be as simple as possible, and the evolution of geological environment can be predicted to a certain extent. The geological barrier can effectively prevent the transfer of radionuclides. The wall rock has enough stretch range.

#### **2. 3 Hydrology and hydrogeology**

The site must be kept away from the water resources protection area, the area where the ground-water can be intruded into, and the area that can be overwhelmed by floods. The hydrogeology features of the site must be beneficial to waste isolation, which include porosity, permeability, chemical composition of groundwater, acidity and basicity, oxidation and reduction, supply and discharge and dynamics of groundwater; and the hydrogeological insulation of the site shall not be damaged because of the excavation of the disposal cave.

## **2. 4 Engineering geology**

The mountain body has good integrity without trenches and gullies, and there is no landslide and collapse topography. The rock body is of uniform integrity, there is no soft stratum in the stratified rock, the karst is not developing, there is no water-bearing structure, and the faults and folds are not developing. The silt and ooze strata, too high hill stress, and active hill areas shall be avoided.

## **2. 5 Social economy**

The site must be kept away from densely-populated areas, the mines valuable for exploitation, the scenic spots, and the natural protective zones. It must be kept far away from the airport, the military testing fields, and the production or storage areas of the inflammable and explosive materials. Water and electricity shall be available without much difficulty.

# **3. The Main Steps and Methods**

## **3. 1 Regional investigation**

According to the siting criteria and the information available, 17 possible sites were finalized for future reconnaissance as a result of the preliminary site investigation.

## **3. 2 Preliminary site investigation**

A comprehensive investigation team was formed with experts in the fields of waste management, geology, hydrogeology, engineering, and environment protection to perform field reconnaissance and collect information on geology, hydrogeology, mine exploitation, environment ecology, and social and economic development. The investigation resulted in preliminary evaluation on the 17 sites and recommendation of several favourable sites for further detailed investigation and analysis.

# **4. The Features of the Recommended Sites in Preliminary Site Investigation**

## **4. 1 Qinshan area**

This area, 8 km away from the Qinshan NPP, is located in the south-east of Haiyan County, Zhejiang Province. It is about 90 km from Shanghai and 78 km from Hangzhou. Qinshan and Fangjia Mountains are linked together, forming an area of about  $10^6 \text{ m}^2$ . The whole mountain area is within the development zone of Qinshan NPP. The population density is  $745/\text{km}^2$ . The highest altitude of the mountain is 185.8 m. It is surrounded by water on the north, east, and south sides, and stretches into Hangzhou Bay. In the west, there is a plain area for agriculture development. The first phase of Qinshan NPP Project is located on the north side of the rear of the mountain. Now, there is an arterial highway directly to Shanghai and Hangzhou. The nearest railway is 26 km from Qinshan and a wharf has been planned for the plant.

This area belongs to Middle China subtropic humid climate zone in the East Asian monsoon climate area. The average temperature is 16.2°C, the maximum temperature, 37.4°C, and the minimum temperature, minus 5.3°C. The relative humidity is normally over 80% and the highest, 98%. The annual rainfall is 1175.6 mm, 167 days, the longest continuous rainfall is 10 days, and the maximum precipitation in one day is 65.5 mm.

The geological formation of this area belongs to Yangzi fault-block area. The west Zhejiang

belongs to earth syncline in Paleozoic Era. Because of Indochina and Yanshan movement, the earth crust suffered from strong pressing, the earth syncline returned to folds, thus the mountain formed. The Paleozoic Era formation laid the basis of this area. By the time of late Jurassic Period, a large scale of volcanic eruption and magma intrusion occurred. The volcanic rock layers built up the covering strata of this area. The layers which can be directly observed in the surface are mainly the second lithology section of late Jurassic Period ( $J_2$ ), volcanic rubble rocks; the upper cover layers are eluvium in Holocene Series ( $Q_1$ ) and Upper Pleistocene Series ( $Q_3$ ). There are some faults through the horseshoe valley near Shuang Long Gang, but they belong to secondary small structures and they are all filled and cemented.

There are not any active faults which may result in earthquake over 5 on the Richter scale within 8 km area around Qinshan NPP. The active faults, which may cause large relative displacement, are not found in the surface or near the surface. The earthquake activity in this area is so weak that the earthquake relative to motive fracture can not happen.

There are mainly two types of groundwater in Qinshan area: (1) the perch groundwater of upper layers, which is normally distributed at contact zone between eluvium of slope section and bedrock. It is characterized by constant changes of water level and small water quantity, which is controlled by atmospheric precipitation; and (2) the fracture water of bedrock with the dense and hard lithology. There is not much fracture water at upper weathering fracture zone or local structure fracture zone. The groundwater is difficult to remain in the deep complete rockbody. It belongs to groundwater micro-aquifer. The surface runoff and the groundwater all flow to seabeach area.

The north, east, and south sides of Qinshan area are surrounded by Hangzhou Bay. There is a plain area in the west of Qinshan and the rivers are interwoven to facilitate water communication. The river adjacent to the NPP is the Changshan River, 8 km from the Plant, and it is the main recharge water source of the Plant. The tidal current in the Hangzhou Bay runs strongly, and the tide range is large. Ganpu tide section, 9 km upstream from the Plant, is the largest tidal range (8.93 m), and the annually-averaged tidal range is 5.47 m. The water in Hangzhou Bay is of low salinity— $10 \sim 22\%$ . The groundwater quality of the seabeach is the same as that of the sea water.

#### 4.2 Daishan lead-zinc mine

The Daishan Lead-Zinc Mine is located in Siping Town in the northeast of Qushan Island, Daishan County, Zhejiang Province. The mine faces the water with hills behind. The east of the island is East Sea, the west is the mouth of the Qiantang River, the south is Daiquyang, and the north is Huangzeyang. It is 120 km from Qinshan. The area is about  $50 \text{ km}^2$  and it has a population of 60,000. The communication to the outside is mainly sea-route. There are wharfs for passengers and oil transportation with a capacity of 1 kt and roads through this island. In the west, 2 km from the mine area, there is a 400 households village with a population of about 1,600, and a small ridge, between the village and the mine area.

The annual precipitation is 899.5 mm; the annual evaporation is 1476.6 mm; the annual relative humidity is 70%. The annual average temperature is  $16.2^\circ\text{C}$ . Typhoon totalled up to 120 times from 1954 to 1981, mostly from July to September. The strong typhoon accounted for 17.5%.

The geologic formation belongs to metamorphic rock of Andeitic lithology section of Chencai

group. The bedrock is well insulated, and separated from sea water. There are two intermediate pits in the mining area, 1,500 m long, and the cross section is  $2.2 \times 2.5$  m in general with maximum one  $4 \times 5$  m. The stoped-out area is about 160,000 m<sup>3</sup>. The rock in the pit is plagioclase hornblende, marble, biotite plagioclase gneiss, hornblende plagioclase, hypergene, granulite, etc. The rock is of good integrity and stability in the mining districts. Though there are no supporting facilities in the stoped-out area, collapse, roof fall, and landslide never occurred. The first intermediate pit is about 50 m above sea level and the second, about 5 m above the sea level.

The fracture water is not developing, so it is dry in the pits.

There is no natural water source or stream in the open air within 1 km in the mining area. The area totals 12,359 m<sup>2</sup>. There is a 300 t wharf in the mine area. The mine was closed in 1976. The island is an area of weak earthquake with scattering frequency.

#### 4.3 No. 771 mine area

The No. 771 Mine is situated at the south-western end of the south-eastern Jinqu basin edge and above the erosion basis. The elevation of Baixi pit mouth is 178 m. The rock of Baixi pit belongs to massive flowline tuff in Moshi mountain groups of Upper Jurassic System, and it is integral and stable.

A group of small-angled and west-eastward overthrust faults are made up of Jiangshan-Shaoxing fault zone in the northern mine area and it is overthrusted on the Moshi mountain group of Upper Jurassic System, therefore, this group is slightly destructed.

The No. 771 Mine is an area of weak earthquake with scattering frequency. In this area, the annual average temperature is 17.3°C, and the annual rainfall is 1717.6 mm. The rainfall is mainly from April to October. There are 153.8 rainy days each year. 5 floods occurred from 1950 to 1980. The landform is relief and deeply dissected to facilitate drainage and prevent floods. The Shengtang River flows through the west side of Baixi pit and its maximum flowrate is 22.44 m<sup>3</sup> per second.

Fracture in Baixi pit's rhyolite is not developing, effective waterproof strata exists. Based on observation that water-bearing faults in No. 20 pit are not developing, the rock has become steady and the compressive strength ranges from 456 to 1,250 kg/cm<sup>2</sup>, the bending strength, from 136 to 470 kg/cm<sup>2</sup>.

The No. 2 fault, which controls the Baixi pit groundwater flow, is composed of clay, kaolin mixed with sandy clay, and debris. As can be seen in No. 20 pit, this fault itself contains no water and has good performance of water-isolation.

The area around the Baixi pit has a sparse population, only about 202 inhabitants living within 2 km range. Clearly, the environmental isolation is really fine.

In Baixi pit, the cross section of the main tunnel is  $2.2 \times 2.5$  m and the length, 500 m and mining fields of branch tunnels are small and irregular, so there is only a limited space available for waste disposal, it is required to excavate new disposal tunnel and gallery.

#### 4.4 Longzhu mountain lead-zinc mine area in Linhai County

Longzhu Mountain Pb-Zn Mine area is 60 km from Sanmen Jiantiao Port and 250 km from Hangzhou. There is a 1,000 t civil wharf at the Jiantiao Port, communication is rather

convenient there. There are only ten households about 50 inhabitants living in this area and it is relatively isolated from the outside world.

This area is within the subtropical oceanic climate circle, the annual average temperature is 17°C, annual precipitation is 1,700 mm, plum rains often fall in spring, the annual evaporation is 1,020 mm, and the annual maximum precipitation, 2242.8 mm.

The geologic formation is grey volcanic tuff which belongs to Moshi Mountain group of Upper Jurassic System. The rock is dense and hard, generally silicified.

The mountain slope of the mine area is steep and the water drains fast. A stream runs through the area. The mine body is above the erosion datum plane. There is no obvious runoff area in the pit, the fracture water has little impact on the water in ore deposit.

The stope is divided into four levels, the total stoped-out area is 220,000 m<sup>3</sup>. The No. 1 middle pit is 247 to 250 m above the sea level, the stoped-out area is about 28,000 m<sup>3</sup>. The No. 2 middle pit is 215 m above the sea level, the stoped-out area is about 56,000 m<sup>3</sup>. The No. 3 middle pit is 182 to 198 m above the sea level, the stoped-out area is 84,000 m<sup>3</sup>. The No. 4 middle pit is 130 to 169 mm above the sea level, the stoped-out area is 53,000 m<sup>3</sup>. The rock in each section is of good integrity and stability. There is no supporting facility in the stope, roof fall and collapse never occurred. The cross-section of the pit is 2×2.5 m ~ 3×3 m.

The earthquake intensity in this area is less than 7 on the Richter scale. It is a stable area as a whole.

#### 4.5 Gaoyubajiaotang

The Gaoyubajiaotang is located in the northwest of Anji County, Zhejiang Province, and its western neighbor is Guangde County, Anhui Province. It is 40 km from Anji County, 120 km from Hangzhou, 220 km from Shanghai, and 200 km from Qinshan. Gaoyubajiaotang is an extension of Xuan-Lang-Guang mound. The rear of the mound is round and plain, and appears as stripped structure. It is a sparsely-populated area.

The Gaoyu area belongs to north subtropic monsoon climate zone, and it is warm and moist there. The annual temperature is 15.6°C, the relative humidity is 80%, the annual precipitation is 1,290 mm, and the maximum precipitation is 1766.6 mm. The rainfall occurs mainly from April to June. The evaporation is 800 mm. The rainstorms appear 37 times during one year, when the rainfall in one day is equal to or more than 50 to 100 mm and the maximum is 100 to 200 mm. The rainstorms mainly appear in September. The typhoon totals 0.8 time in one year, mainly from August to October.

This area belongs to Tai Lake hydrographic net. In the northeast 10 km away from the Bajiaotang area, there is a Tianzigang Reservoir with a capacity of 18,000,000 m<sup>3</sup>.

The groundwater belongs to porous fracture water of weathering red bed. The aquiferous formation is weathering rock of the Qifang Village group of Cretaceous System and the main component is clay with poor permeability, which leads to an extreme shortage of water. The water capacity in the civil wells is 12 to 13 tones per day. The groundwater dynamics is in an unstable state. The groundwater level is 10 m subsurface. The groundwater is only recharged by rainwater and flows steadily to surrounding ditches, i.e. local recharge and local discharge. The surface water is the drinking water source for the inhabitants.

The surficial stratum in this area belongs to Guangde group of Upper Jurassic System and Qifang Village group of Lower Cretaceous System.

The bed rock is argillaceous sandstone.

This area is located along the central line of the fourth small belt of the east-west tectonic system, which belongs to later Yanshan movement. Faults and fractures might be developed parallel to the edge of the basin. They are small in scale and the rock is subject to least destruction.

This area is of weak earthquake with scattering frequency. The main plant in this area is oil-tea camellia. It is rich in bentonite resources there. The average population is about  $230/\text{km}^2$  inhabitants per square km.

## 5. Preliminary Assessment of the Recommended Sites

According to the above-mentioned characteristics of the different sites, it is regarded that Qinshan as cave disposal site for Qinshan NPP low- and intermediate-level radwastes has such advantages as stable formation, simple hydrogeology conditions, good isolation properties, and convenient transportation. There are few township enterprises within 5 km of Qinshan NPP, and there is no large enterprise within 20 km. However, the Qinshan area is part of Hangjiahu plain of Shanghai economic development zone, a developed area in agriculture, fishery, and light textile industries. The essential factors unfavourable to the siting determination are: the long-term development project in this area, the distribution of the environmental radiation dose of the Qinshan long-term nuclear power project, the development of north-south lake tourism area 12 km from the plant.

As to Daishan Lead-Zinc Mine, the bedrocks have good water isolation. Observed from the pit, the fractures are not developing, and the tunnels are dry. There is a 400 households village nearby, separated by a small hill. The mining area faces the water with hills behind, and is well isolated with human activities. The abandoned mine has certain spacial capacity, and there is potentiality to expand the capacity of the mountain body. Although the mining area is closed now, there are some houses and technical workers there. There is a wharf in the mining area, it is 120 km from Qinshan by sea route, the sea transportation is very convenient. But the sea area of the Qushan island where the mining area is located is a main part of the Zhoushan fishing ground, the public psychology and social opinion will possibly cause unfavourable conditions relating to development of such a disposal site.

The Baixi pit of No. 771 Mine will be decommissioned soon, the equipment remain complete and there are enough technical workers. The mountain body is under good integral and stable conditions. But the existing excavation space is small and irregular, the abandoned pit can hardly be used as a whole. The main disposal space needs to excavate. The communication in the mining area is convenient, though it is a bit too far from Qinshan.

The Longzhu Mountain Lead-Zinc Mine area is located in remote mountains of Linhai County. There are very few inhabitants there, and the separation is good. The bedrock of this mine belongs to volcanic tuff. The rock is integral and stable, the fractures of the bedrock is poor, and the pit is dry. The tunnels are long and the excavation space is large, the availability in space is high. But the reconstruction is complicated. The mining area is about 360 km from

Qinshan highway, and the communication wants land- and water-coordinated transportation.

The Gaoyubajiaotang area is 200 km from Qinshan. The communication is convenient. The population density is relatively low. As a state-owned barren area, it is easy to make a requisition of land. The surface water in this area is well drained out, and the ground water is 12 m below the surface. But the bedrock is gritrock, the separation property of the strata is poor, the demolitions are required for the shallow strata disposal. This area is part of the edge area of Hangjiahu plain, and there is potentiality for further exploitation.

The above sites all have the prerequisites to dispose of the low- and intermediate-level radwastes. However, the assessment of the site characteristics shall take a lot of practical field survey so as to gain more specific and detailed data and go deep into the analyses.

## **SESSION IV**

### **SCIENTIFIC RESEARCH AND APPLICATION**

# USE OF A TANGENTIAL FILTRATION UNIT FOR PROCESSING LIQUID WASTE FROM NUCLEAR LAUNDRIES

X. Augustin A. de Buzonnière

TECHNICATOME

H. Barnier

Commissariat à l'Energie Atomique

CEA CADARACHE - DSD/SEP/SEATN

## ABSTRACT

Nuclear laundries produce large quantities of weakly contaminated effluents charged with insoluble and soluble products.

In collaboration with CEA, TECHNICATOME has developed an ultrafiltration process for liquid waste from nuclear laundries, associated with prior insolubilization of the radiochemical activity.

This process "seeded ultrafiltration" is based on the use of declogable mineral filter media and combines very high separation efficiency with long membrane life.

The efficiency of the tangential filtration unit which has been processing effluents from the Cadarache Nuclear Research Center (CEA-France) nuclear laundry since mid-1988, has been confirmed on several sites.

## **Introduction**

Contaminated liquid waste used to be decontaminated by processes band either on evaporation process or on frontal filtration. The evaporation is a rather expensive technique for low-level liquid waste with activities only slightly above the limit for unrestricted release. Detergent foaming can also affect evaporator operation. The frontal filtration without regeneration of the filter surface causes... substantial production of secondary solid waste comprising charged filter elements.

Cross-flow filtration, and membrane-type ultrafiltration in particular, avoids irreversible clogging of the filter media and their consequent disposal.

In cooperation with the CEA, TECHNICATOME has developed this process with the following objectives:

- concentration of the radiochemical activity within a limited volume of waste, stockable after solidification treatment and
- achievement of a decontamination factor high enough to allow release of the filtrate without further treatment.

## Liquid Waste from Nuclear Laundries

Radionuclides including Co, Ag or Cs are routinely present in slightly contaminated liquid waste while others such as Ru, Sb, Cu or Hf are occasionally detected with ranges of activity between 200 Bq/l to 10 000 Bq/l.

Usually such waste includes suspended solids (about 1 g/l maxi) and anionic detergents (about 0.1 g/l). This type of detergent forms complexes with radioactive cobalt in liquid waste. Besides, additions of sodium hypochlorite may be necessary in interim storage to prevent bacterial growth.

### Description of the Adsorption-Ultrafiltration Process

This process involves partial on-line batch treatment.

#### The first step is to make the radiochemical activity insoluble

The waste is placed in the treatment tank for initial insolubilization; chemical additives and mineral-type adsorbers are introduced simultaneously into the waste, and thorough mixing is obtained by a few minutes stirring.

#### Separation of the insoluble matter is carried out in an ultrafiltration loop

The mixture is transferred to a filtration loop in which it revolves at a high speed inside tubular banks of mineral membrane filter elements. As a result of the differential pressure between the inside and outside of the filtration tubes, part of the purified liquid is passed through the filter media. The rest is recycled to the treatment tank, and concentrated progressively as suspended matter as filtration proceeds.

The concentrate collected is stocked prior to solidification. After radio-chemical measurement, the filtrate is evacuated to the release system, except in the event of an incident.

The process is illustrated in Fig. 1.

The ratio of treated waste volume to residual concentrate volume gives the Volume Reduction Factor for the process. This factor is closely related to the geometry of the available area and to the technology of the system (pumping device, piping layout, etc.). It may be improved by incorporation of a separator in the recycling loop.

The operational parameters (circulation speed in the loop, pressure drop, etc.) are selected during preliminary tests on pilot plant in order to determine:

- average filtration flowrate and
- minimum clogging of the membranes, and periodicity for chemical flushing to restore membrane permeability.

## Membranes

The use of a ceramic filter layer (see Fig. 2) gives membranes with:

- very long life which can be up to around ten years, and
- exceptional chemical resistance, allowing initial permeability to be integrally restored by chemical cleaning under extreme pH conditions.

These membranes are currently in increasingly widespread use, in particular in the agro industries (dairy cheese, beverages), biotechnology, pharmaceuticals, automobile industry, etc.

### Application Example: Treatment of Radioactive Wastes from a Nuclear Research Center Laundry (CADARACHE-FRANCE)

#### Characteristics of waste

The main characteristics of the waste from the clothing laundry are as follows:

- total suspended solids: 1 g/l (mean),
- detergent content: 0.1 g/l, and
- global radiochemical activity (gamma spectrometry): 3,500 Bq/l (mean), mainly due to the following radionuclides: Co-58, Co-60, Cs-137, Ag-110m, Mn-54.

#### Treatment unit

The capacity of the unit is 12 m<sup>3</sup> of liquid waste per week in 80 to 120 hours.

The ultrafiltration unit used is equipped with mineral membranes.

The ionic species, which are either soluble or chelated by the active surface agent, are fixed on a mineral adsorber prior to ultrafiltration.

#### Treatment performance

The average activity of the filtrate is about 30 Bq/l, i. e. substantially below the release limit in France (370 Bq/l before dilution). The filtrate volume is more than 90% of the initial volume of the waste.

The average filtering flowrate achieved is approximately 100 l/hm<sup>2</sup> under nominal operating conditions.

When the weekly production of liquid waste have been treated, the initial permeability of the membranes is restored by means of an acid-base chemical heat flushing process. This flushing generates a small volume of very slightly contaminated liquid (approx. 30 Bq/l) which is either released with the filtrate, or recycled to a waste tank for treatment.

The quantity of adsorbers used depends on the radiochemical activity of the waste, but is always very low.

A schematic diagram of the process is shown in Fig. 3.

### Nuclear Laundry for a PWR Power Station

Application of this technique to the liquid waste of the laundry for weakly contaminated clothing at a PWR power station has produced similar results into similar equipment.

#### Characteristics of the effluent processed

- suspended matter: 30 mg/l to 1 g/l,
- detergents: approx. 0.5 g/l,
- pH: 7 to 8.5, and

- radioactivity: 500 to 1,500 Bq/l.

### Performance

- elimination of all suspended matter from the effluent,
- reduction of the radioactivity of the effluent to less than 28 Bq/l,
- concentration factor by volume between 36 and 108, and
- residual concentrate heavily charged with suspended matter (30 to 110 g/l), with global radiochemical activity of about 20,000 to 100,000 Bq/l.

### **Conclusions**

The performance described above has been confirmed by several campaigns carried out on several real wastes.

The adsorption-ultrafiltration process is not limited to small quantities of waste. The modular design of the filter elements makes it possible to install units with large membrane surface areas. For instance, a unit incorporating 4 modules each with 252 filtration tubes (i.e. approximately 20 m<sup>2</sup> of membrane surface) can treat 190 m<sup>3</sup> of waste/week.

Moreover, R&D works have proved the possibility of phosphate elimination with this process.

### **Other Applications**

The ultrafiltration process, after insolubilization if necessary, can be used for volume reduction for a wide variety of radioactive wastes.

Indeed, the use of high chemical and thermal resistance mineral ultrafiltration membranes, together with a judicious choice of insolubilization agents, makes it possible to treat waste containing a large variety of chemical and radiochemical elements.

These include the following liquids:

- used effluents (floor drains, chemical liquids, residual liquids),
- decontamination liquids,
- storage pool water, and
- carbonated effluents (solvent regeneration).

The versatility, operating principle (filter chemically regeneratable in situ, self-regulated declogging, easy automation and limited maintenance) and performance of this process, make it particularly suitable for applications in the nuclear industry.

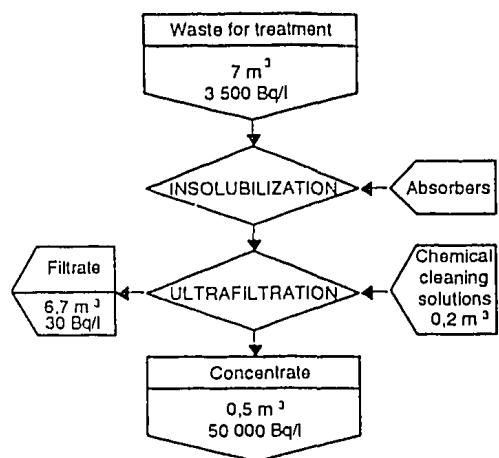


Fig. 1 Adsorption-Ultrafiltration Applied to Laundry Waste Flow-Sheet

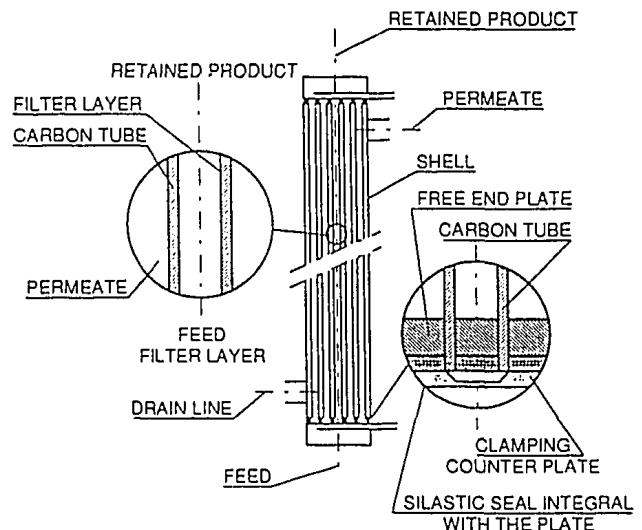


Fig. 2 Ultrafiltration Module-Principle

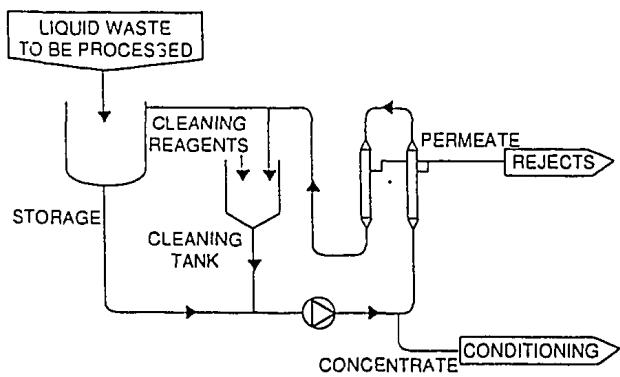


Fig. 3 Schematic Diagram of a Ultrafiltration Loop

# A STUDY ON FREE RADICAL OXIDATION OF SPENT RADIOACTIVE ION-EXCHANGE RESINS

GENG Zuohong WU Jiaquan  
YUN Guichuan WU Tianbao

Institute of Nuclear Energy Technology,  
Tsinghua University

## ABSTRACT

The resin dissolution process was conducted successfully in bench-scale tests. The polystyrene based strong acid cation-exchange resins with water content of about 58% (wt) were dissolved by hydrogen peroxide and ferrous ions as catalyst under pH of 2.0~3.0 adjusted by sulphuric acid. For the same objective of dissolution of the strong basic anion-exchange resins with water content about 63% (wt), citric acid was the best choice for pH control, and the use of  $\text{Fe}^{2+}$  and  $\text{Cu}^{2+}$  was verified having a synergistic effect. Mixed resins were also dissolved successfully under proper conditions. The dissolution temperature was generally below 99°C. The COD and TOC levels of the dissolution residues depended on the doses and dosing rate of hydrogen peroxide as well as the catalyst supplied. All the three types of dissolution reactions gave the similar degradation pattern.

## 1. Introduction

Spent radioactive ion-exchange resin (IER) is one of the main wastes arising from nuclear installations. Although the amount of resins produced in batches may not be large, many difficulties exist in solidification and safe disposal because of the organic composition and granular or powdered shape of resins. Solidification in status quo also causes problems such as an increase of ultimate disposal volume, poor stability for long-term disposal, etc.

Spent IER should be treated into a suitable form for the sake of safe and economic final disposal. As a kind of organic waste, it can be converted to inorganic waste which may be easily disposed of by incineration pyrolysis or high temperature wet oxidation. But for treatment of radioactive ion-exchange resins, it's extensively regarded as a good way to dissolve IER with free radical chemical oxidation, using hydrogen peroxide as oxidant, which could be conducted under atmospheric pressure and comparatively lower temperature.

The objectives of the study are to:

- (1) Demonstrate that the dissolution of IER can be carried out by catalyzed wet oxidation — free radical chemical oxidation; and
- (2) Investigate the operation conditions of the dissolution process in bench-scale.

## 2. A Brief Introduction to Process

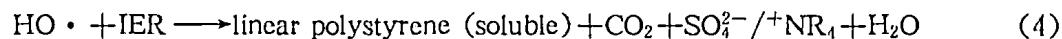
The IERs dissolved here are sulphonated and/or aminated cross-linked polystyrenes which are connected to form three-dimensional network polymers by divinyl benzene. The structures of such IERs are presented in Figs. 1 and 2.<sup>[1]</sup>

The mesh structure of IER makes itself more stable.

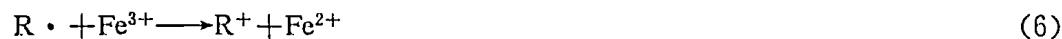
$H_2O_2$  is commonly used as a source of hydroxyl radical ( $HO \cdot$ ). It is suggested that the decomposition of  $H_2O_2$  catalyzed by single-electron reductants such as iron (II) and copper (I) ions may generate  $HO \cdot$ .<sup>[2]</sup> The reaction between hydrogen peroxide and ferrous sulphate is the typical radical reaction discovered by Fenton in 1894, which Haber and Weiss described later in the following chain reactions:<sup>[3]</sup>



Hydroxyl radical with very high reactivity could react with organic substrates either by hydrogen abstraction or by addition to an unsaturated system. Such reactions may be used in both organic synthesis and organic degradation. When used in IER dissolution, it can be represented as the following reactions:



The reaction (4) and (5) involve organic free radicals ( $R \cdot$ ), which may be oxidised by high valent ions such as  $Fe^{3+}$  and  $Cu^{2+}$ , as shown in the following reactions:<sup>[4]</sup>



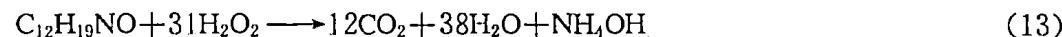
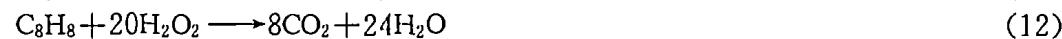
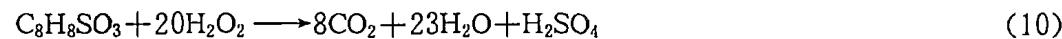
The production of  $Cu^+$  can increase the catalysis efficiency of  $Fe^{2+}$  on account of existance of the following reaction:



The reactions (1)~(8) form an effective chain cycle, through which the high polymer IER is finally degraded to simple organic substances with low molecular weight. It was also suggested by Haber and Weiss that the chain cycle could be terminated by the following reaction:<sup>[3]</sup>



The ideal results of the resin dissolution might be presented as follows:



Equations (10) and (13) represent the dissolution reactions of the part of IER to which function groups are attached, while equations (11) and (12) stand for the reactions of cross linking agent and styrene unit separately.

### 3. Experiments

#### 3. 1 Material and equipment

Wet fresh particulate resin with moisture content 58~63%, that was abstracted from resin slurry, and 30 vol% hydrogen peroxide were chosen in the study. In addition to those

reactants, 0.10 M  $\text{Fe}^{2+}$  and/or 0.10 M  $\text{Cu}^{2+}$  solutions and 10 vol% sulphuric acid and/or citric acid were fed as catalyst and acidulant respectively.

All the small-scale experiments were performed in a 500 ml four necked glass flask equipped with a water condenser and the flask was immersed in an oil bath to keep the desired reaction temperature. The vent line was sealed to analyze gas products exactly and accurately. Here, an electromagnetic mixing heater was also used to provide mixing and heating. A sketch of the laboratory resin oxidation system is shown in Fig. 3.

### 3.2 Process description

A 25 g batch of wet resin was added into the flask firstly, sulphuric acid and/or citric acid was then used to adjust pH value of resin slurry to 2.0~3.0. A small amount of catalyst should be fed before the reaction to accelerate the access to oxidation process.

When temperature in the flask rose to 80 °C, the oxidation reaction was available, the resin beads began to become deep red steadily, and some of them were turned to be soluble. With the addition of  $\text{H}_2\text{O}_2$  and catalyst, more and more resin beads were dissolved and a large amount of gas products was released from the main reaction body. An hour later, nearly all the beads became a deep red solution, and gas products, including  $\text{CO}_2$  and  $\text{O}_2$ , were continuously formed. At the tail stage, the solution was becoming light yellow, the amount of  $\text{CO}_2$  produced was decreased, while  $\text{O}_2$ , the direct-decomposition product of  $\text{H}_2\text{O}_2$ , was increased greatly. By the end of the reaction,  $\text{CO}_2$  was almost no longer generated.

### 3.3 Chemical analysis

From observation, the reaction process could be divided into two stages. The overall pattern of the reaction is as following:

- (1) At the first stage, cross-linked polystyrene was oxidised to linear soluble polystyrene, as represented in equation (4). It was clearly observed that the original resin beads were converted to a deep red solution with high organic content. The release of  $\text{CO}_2$  was substantial, while the  $\text{O}_2$  evolution was very low. (see Fig. 4); and
- (2) At the second stage, linear polystyrene was further degraded to simple organics such as aromatic acids, as represented in equation (5). As observed, the solution was becoming more and more light and clear, and the crossover of  $\text{CO}_2$  and  $\text{O}_2$  produced began, the release of  $\text{O}_2$  was dropped nearly to zero if no  $\text{H}_2\text{O}_2$  added. (see Fig. 4)

As a result of fairly complete dissolution of resin, the residual solution was generally composed of simple aromatic acids, sulphuric acid, and inorganic salts, and also amines if there was a dissolution of anion-exchange resin. The pH value of the residual solution was often lower than 1.5. In some cases, a very few of solids, which were not easily degraded, were also left in the final solution.

On an average, the off gas consisted of 40%  $\text{CO}_2$ , 40~50%  $\text{O}_2$ , some water, and a small amount of evaporable organics, while, in the condensate apart from predominated water, organic acids, amines, and other organics of low molecular weight existed. The total organic carbon (TOC) level of the condensate was about 2000 mg/l, and the pH value was approximately 3.0.

#### 4. Results and Discussion

More than 50 bench-scale studies and a series of orthogonal tests were performed to investigate the oxidation process conditions and determine the appropriate parameters of dissolution of the three types of resins. The results demonstrated that there were many factors having influences on resin dissolution, such as the amount and the manner of H<sub>2</sub>O<sub>2</sub> addition, catalyst feed rate and mode, temperature, pH value and so on. The following discussion gives a good view of the important factors.

- Temperature

Resin dissolution reaction is highly sensitive to temperature. At the room temperature, reaction system has no change, even if H<sub>2</sub>O<sub>2</sub> was overfed. The experiment showed that at a low temperature, direct-decomposition of H<sub>2</sub>O<sub>2</sub> was the prevailing reaction regardless of the very slow reaction rate, and resin oxidation couldn't be visible unless the temperature rose above 80°C. Thus, the initial temperature of dissolution reaction should not be below 80°C.

Obviously, the higher the temperature is, the faster the reaction will be (see Fig. 5). But for resin oxidation, too high temperature may cause a lot of troubles, such as foaming, diffusing of nuclides to gas phase, etc. Therefore, the desired temperature is around 95°C, generally between 90 and 99°C, under the initial boiling point.

- Hydrogen peroxide

Hydrogen peroxide, as one of the reactants, is a key factor affecting dissolution process. Firstly, the amount of H<sub>2</sub>O<sub>2</sub> added often has significant contribution to reaction results. The more H<sub>2</sub>O<sub>2</sub> added, the more completely resins oxidised. This can be obtained from Fig. 6. But too much consumption of H<sub>2</sub>O<sub>2</sub> certainly makes operation cost very high. Considering the economical benefit of the process, the amount of H<sub>2</sub>O<sub>2</sub> added should be reduced as low as possible, based on the efficiency of H<sub>2</sub>O<sub>2</sub> usage.

Secondly, H<sub>2</sub>O<sub>2</sub> dosing rate gives a great impact on operation process, especially on the reaction temperature. A dosing rate at high level could inevitably increase the temperature sharply, causing foaming and pressurizing so that the operation becomes difficult and unsafe. However, if the dosing rate is too slow, the reactivities may be constrained, and the reaction time becomes very long. Thus, an appropriate H<sub>2</sub>O<sub>2</sub> dosing rate is rather important.

Fig. 7 is the tendency chart of orthogonal test for 25 g wet cation-exchange resin dissolution. The comprehensive assessment index relates to both the degree of resin oxidation which is represented by chemical oxygen demand (COD) level in final residual solution, and the efficiency of H<sub>2</sub>O<sub>2</sub> usage expressed by release factor of O<sub>2</sub> produced. The lower the index is, the better the result should be. It's clearly shown in Fig. 7(a) that, for 25 g wet cation-exchange resin dissolution, 200 ml H<sub>2</sub>O<sub>2</sub> (equal to a dosing rate of 0.7 ml/min and 5 hours run time) is the optimal dose.

- Catalyst

The bench-scale studies prove that ferrous ion is a highly effective catalyst in dissolution of cation-exchange resin, and for anion-exchange and mixed resins, the use of Fe<sup>2+</sup> and Cu<sup>2+</sup> is preferable to any single catalyst because of their synergistic effect on the free radical oxidation.

The experimental results also indicate that the amount of catalyst and feed mode have significant effects on the decomposition of ion-exchange resins. Generally, the more catalyst feed, the

more complete decomposition and consequently the higher operation cost. On the other hand, the various feed modes can lead to different results, because some reaction products such as aromatic acids may develop complex reaction with metal ions and hinder hydroxyl radical chain reaction. Continuous feed mode is better than either premixing or intermittent one.

For dissolution of 25 g wet cation-exchange resin, 12 ml 0.1 M  $\text{FeSO}_4$  solution is preferable, as shown in Fig. 7 (b).

- pH Value

The wet resins should be acidulated before catalyst feeds to prevent coating of iron hydroxides precipitating on the surface of the resin beads. However, very low pH value conditions cause the efficiency of  $\text{H}_2\text{O}_2$  usage to be poor, and increase base addition in the subsequent neutralizing treatment. So the pH values of the resin slurry are usually adjusted to 2.0~3.0, just below 3.4, the pH condition of precipitation of iron hydroxides.

Sulphuric acid is used as acidulant for cation exchange resin. But for anion-exchange resin, citric acid is preferred. The dissolution mechanism indicates that only when the catalyst ions are bound to ion-exchange sites can the resins be well decomposed. Therefore, addition to acidulation, citric acid, a weak chelating agent of  $\text{Fe}^{2+}/\text{Fe}^{3+}$  and  $\text{Cu}^{2+}$ , could convert metal ions to complex anions which are easily bound by the function groups of anion-exchange resins. By this means, the oxidation process of anion-exchange resin is effectively promoted.

As for mixed resins, sulphuric acid and citric acid should be added in order to meet the needs of decompositions of both types of resins.

The test shows that the condition of pH 3.0 is suitable to the dissolution of 25 g wet cation-exchange resin, as shown in Fig. 7 (c).

- Foaming

Foaming is a problem particularly in dissolution of anion-exchange and mixed resins. At high temperature, foaming becomes so serious that the reaction system can be contaminated and pressurized. In order to eliminate the negative consequence, tributyl phosphate (TBP) is used as anti-foaming agent. For bench-scale tests, about 1 ml TBP is sufficient.

- Reaction Time

It's necessary to state that the residual solution, which has been put aside for some time, has lower concentration of organics than that one discharged just from the reaction vessel. Surely, the oxidation process is still proceeding due to the existence of residual  $\text{H}_2\text{O}_2$ . This can be well demonstrated by Fig. 8.

In fact, many factors such as batch size, complexes, mixing, etc., has influences on oxidation process of resin. What's more, the factors may interfere with one another. Therefore, the complex relationship between the various factors should be considered in decision of good process conditions.

## Conclusions

The laboratory tests have demonstrated that the three types of ion-exchange resins can be efficiently oxidised by free radicals produced through metal ions catalyzed decomposition of hydrogen peroxide. The appropriate process conditions and corresponding results are listed in Table 1.

Finally, the residual solution, which contains a predominated percent of radioactive nuclides, needs to be neutralized, and then concentrated to a small volume of solution to be prepared for cementation. A satisfied volume reduction factor is expected to be achieved in final disposal.

In addition, the decontaminated solution containing citric acid might be used to adjust pH value of anion-exchange resin instead of pure citric acid, thus both operation cost and waste volume reduced.

Conclusively, the development of resin oxidation process is in sight on account of the feasibilities in technology and economics.

Table 1 The conditions and results of resin dissolution

Conditions and results	Cation-exchange resin	Anion-exchange resin	Mixed resin (weight ratio 1 : 1)
Pure H <sub>2</sub> O <sub>2</sub> comsumption/ wet resin (kg/kg)	3.52	4.40	4.22
Catalyst fed* /wet resin • run time (kg/kg • min)	A : 4.45×10 <sup>-5</sup>	A : 2.12×10 <sup>-5</sup> B : 1.84×10 <sup>-5</sup>	A : 1.85×10 <sup>-5</sup> B : 1.61×10 <sup>-5</sup>
Temperature (°C)	90~99		
pH value	3.0	1.5	3.0
Acidulant	Sulphuric acid	Citric acid	Sulphuric acid and citric acid
Run time (hrs)	5	7	6
Anti-foaming agent/ wet resin (l/kg)	—	0.04	0.04
Reaction room/wet resin (l/kg)	20		
Other conditions	agitating, nonreflux condensing		
TOC level of final solution (ppm)	100~1000	1000~5000	500~1000
Efficiency of H <sub>2</sub> O <sub>2</sub> usage (%)	50~60		

\* A—FeSO<sub>4</sub> • 7H<sub>2</sub>O B—Cu(NO<sub>3</sub>)<sub>2</sub> • 3H<sub>2</sub>O

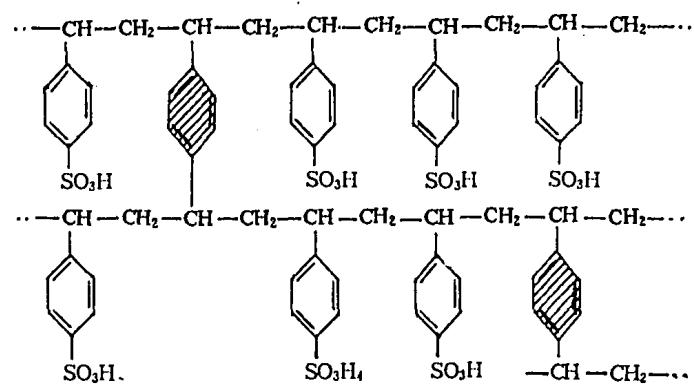


Fig. 1 The structure of strong acid action-exchange resin

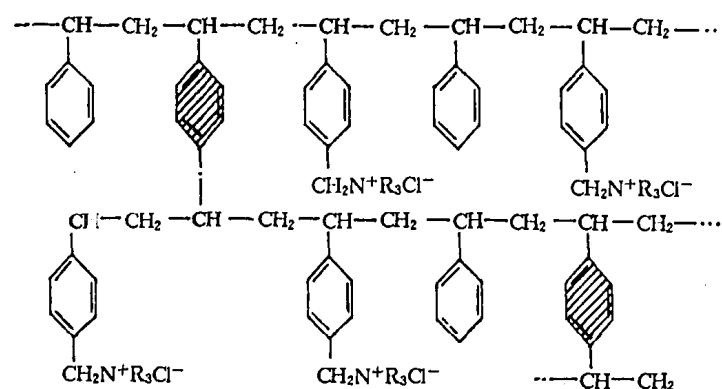


Fig. 2 The structure of strong basic anion-exchange resin

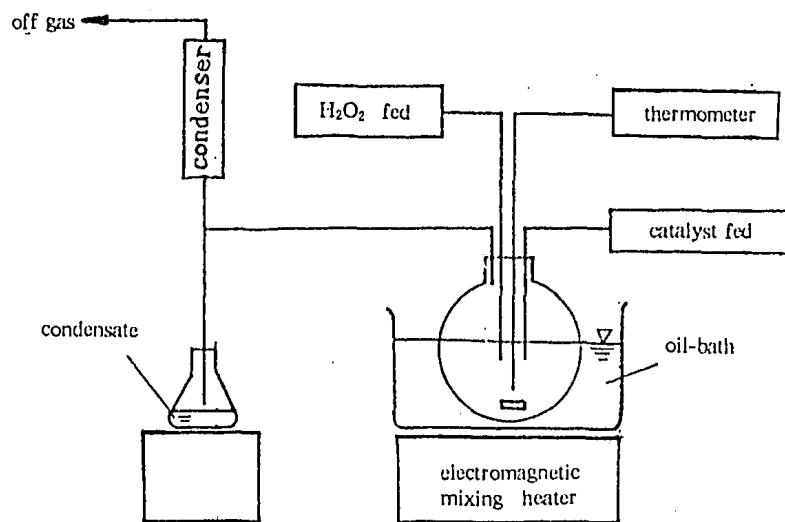
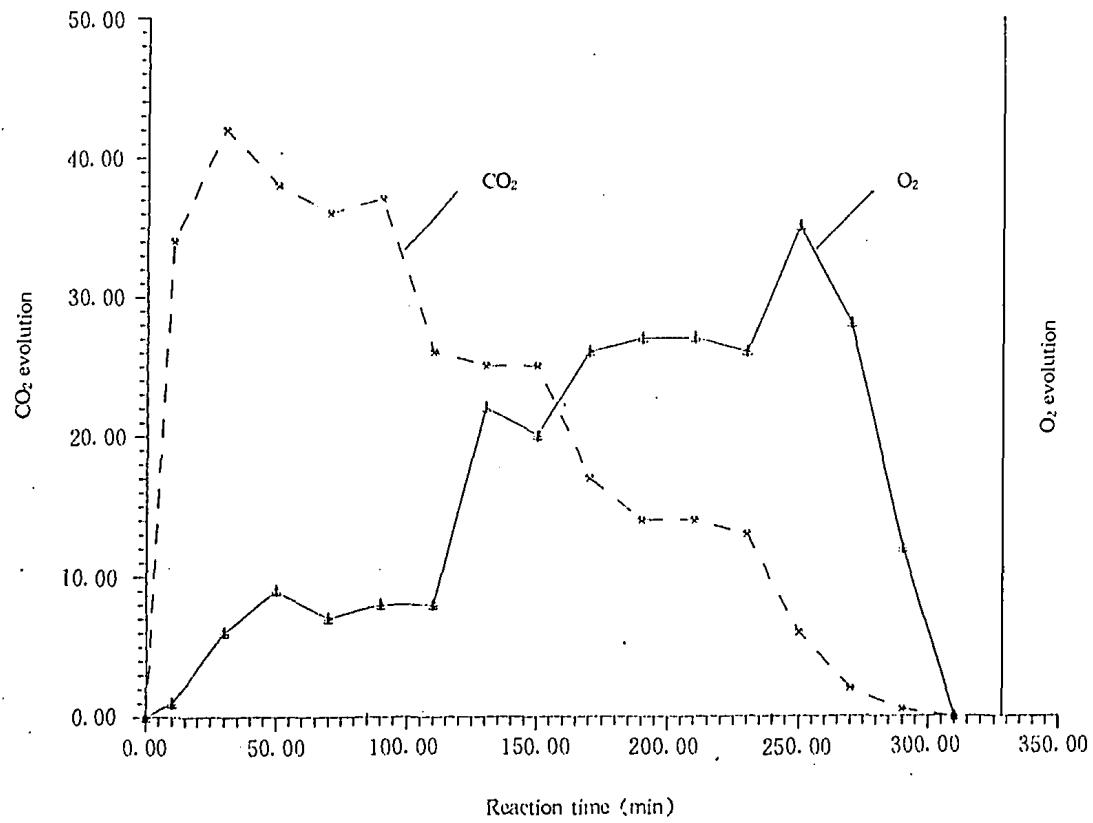


Fig. 3 A sketch of the laboratory resin oxidation system



Note: The evolution of CO<sub>2</sub> and O<sub>2</sub> were measured by GCG.

Fig. 4 The release of CO<sub>2</sub> and O<sub>2</sub> during dissolution reaction

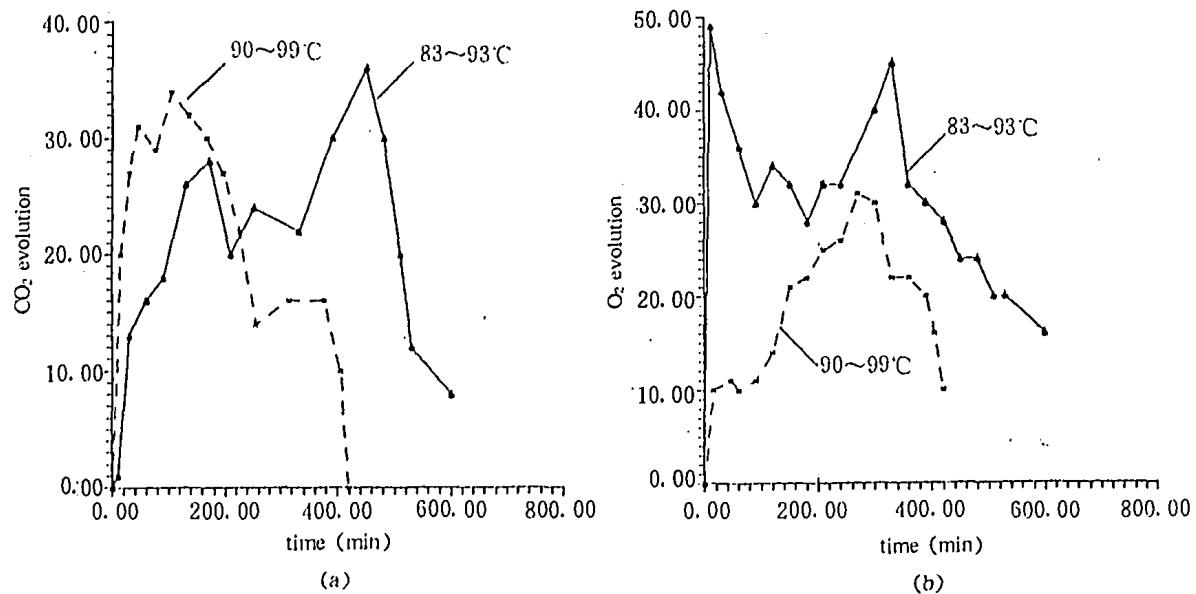


Fig. 5 The effect of temperature on resin dissolution

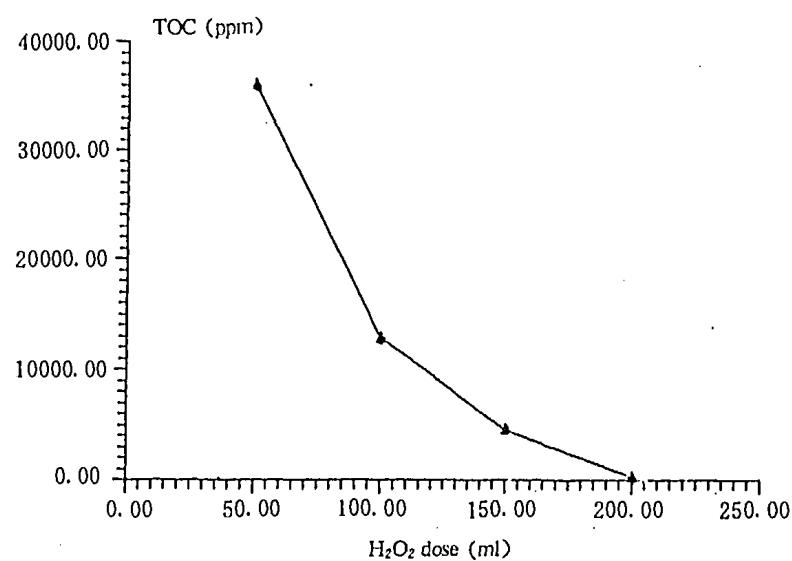


Fig. 6 The relationship between  $\text{H}_2\text{O}_2$  dose and the result of resin oxidation — TOC

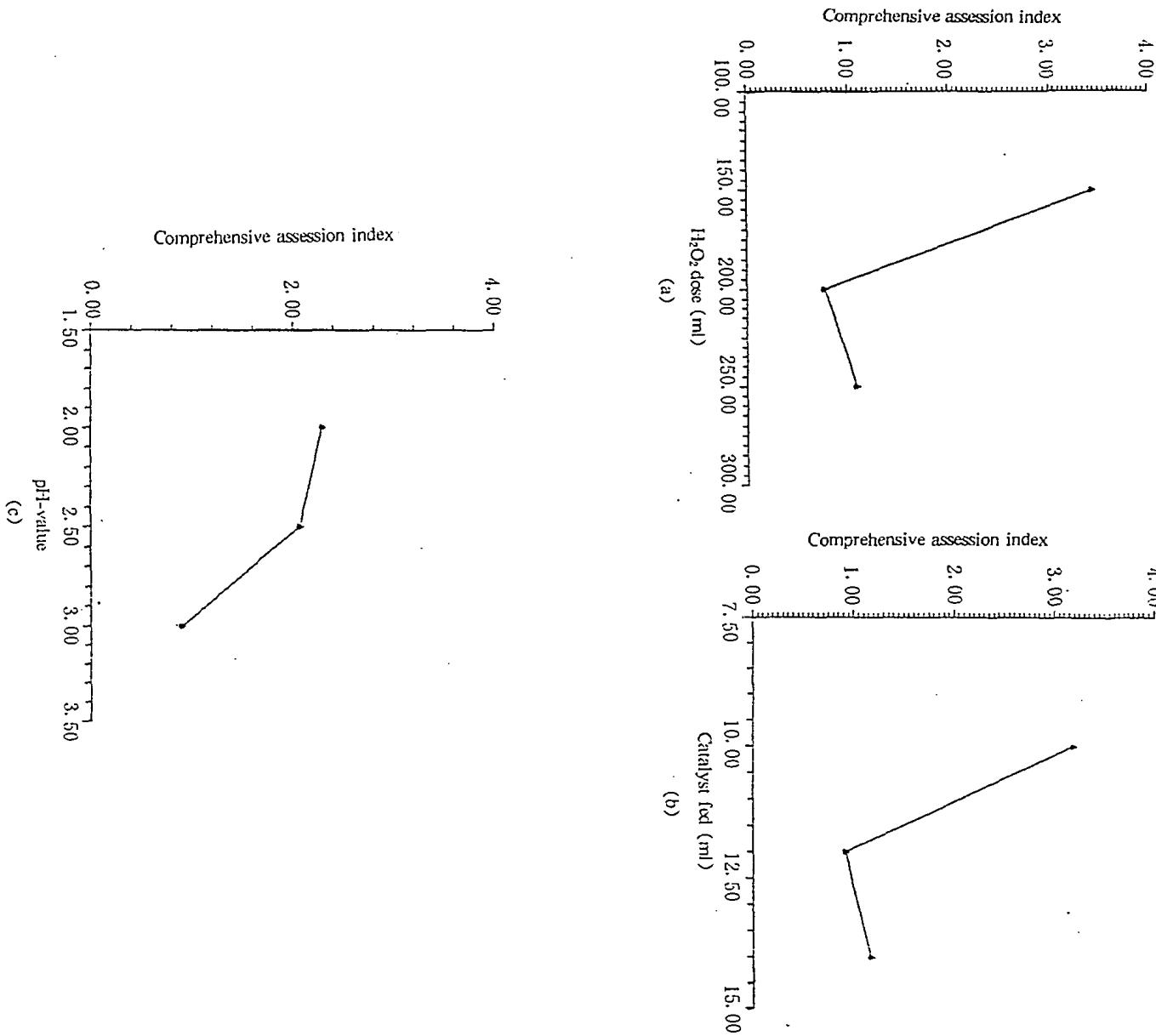


Fig. 7 Tendency Chart

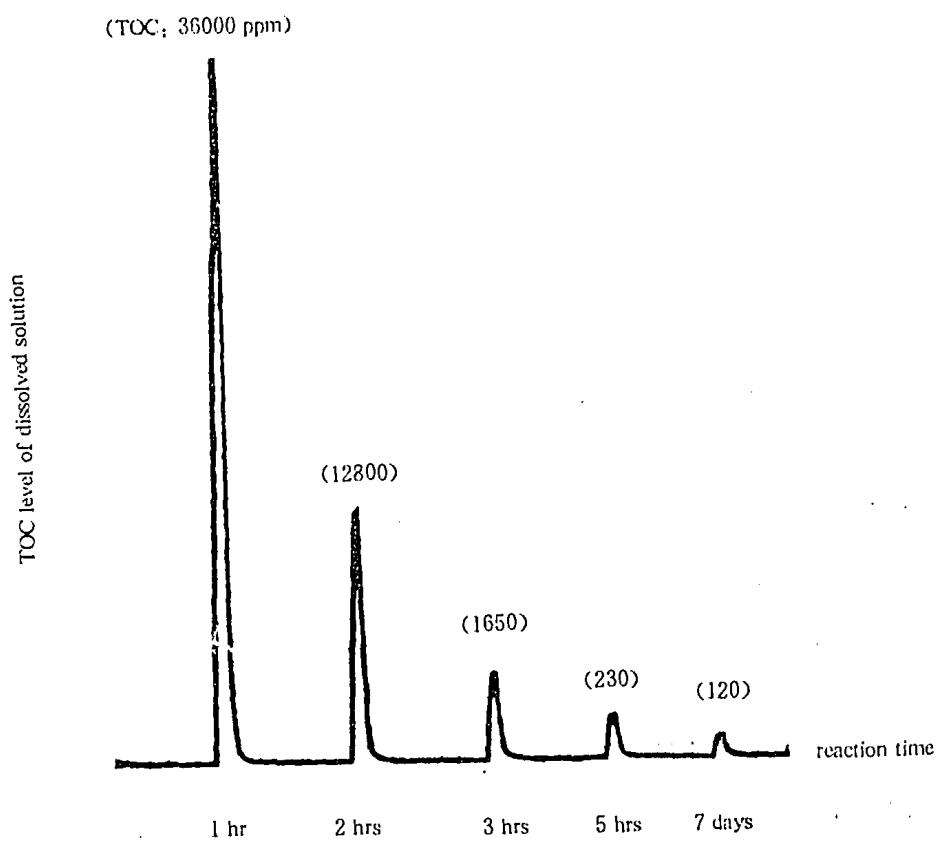


Fig. 8 The relationship between TOC lever of dissolved solution and reaction time

### References

- [1] XIA Duwei, "Ion-Exchange Resin", Chemistry Industry Press, 1981 (in Chinese).
- [2] D. C. Nonhebel, J. C. Walton, "Free-radical Chemistry", Cambridge, at the university press, 1974
- [3] F. Haber, J. Weiss, "The Catalytic Decomposition of Hydrogen Peroxide by Iron Salts". Proceedings of the Royal Society of London, Series A, 1935, 1. 24. London
- [4] Chevs Walling and Shiniik Kato, "The Oxidation of Alcohols by Fenton's Reagent, the Effect of Copper Ion", J. A. C. S. Aug. 25, 1971.

# TREATMENT OF LL AND ML LIQUID RADWASTE

## THE SGN EXPERIENCE

Eric TCHEMITCHEFF      Patrice ROUX  
SGN, FRANCE

### 1. Introduction

The operations involved in the treatment of a low- or medium-level liquid radwaste generated at a nuclear plant include:

- concentration of the radioactivity present into a minimum volume for conditioning and subsequent disposal; and
- the ability to release most of the aqueous phase into the environment after decontamination.

This problem is faced by all the actors in the nuclear fuel cycle, and it is important because it concerns significant volumes.

With the backing of its parent company COGEMA, of CEA, the French Atomic Energy Commission and benefiting from assistance contracts with ANDRA and cooperation agreements with EDF, SGN has built up considerable know-how and enjoys thirty years of feedback in the industrial management of liquid effluents from the entire French nuclear industry, expertise which it offered very early to its foreign clients.

SGN has accordingly completed the engineering and commissioning of many LL/ML radwaste treatment facilities, relying on two major industrially proven technologies: evaporation and chemical coprecipitation.

These units have always been integrated within a concept aiming at the minimization of effluent production at the source, careful segregation of waste, and good engineering of the collection, storage, and sampling systems, intended for the optimum integration of all the links in the treatment chain up to final disposal.

### 2. Evaluation of LL and ML Radwaste

Evaporation is the process by which a solution is concentrated through the removal of a volatile phase by the action of heat.

The liquid radwaste is evaporated to form a small stream containing all the non-volatile components at the column bottom, and a head stream composed mainly of steam, which is condensed, other volatile compounds, and traces of entrained initial compounds.

Liquid radwaste evaporation is the most widespread form of treatment in reprocessing plants, nuclear power plants, and research centres. Combined with an effective steam purification technique, it helps to obtain very high decontamination factors (DF) for nearly all the radionuclides present, and generally makes it possible to discharge the distillates obtained directly into the environment.

SGN, which has built up considerable industrial experience in this technology, that it has been

used extensively for delicate evaporation and distillation operations at the very core of the process applied in reprocessing plants, enjoys complete industrial expertise in evaporators featuring external boilers and natural circulation (thermosyphon evaporators). The simultaneous application of this technology to non-nuclear uses on industrial wastes, where especially difficult problems of foaming and crust formation are encountered, and where the economic factors are extremely important, have enabled SGN to extend its know-how and references to other types of evaporator, better suited to the concentration of LL/ML liquid radwaste, namely :

- "pot" type evaporators, with double jacket or submerged tube bundle, and
- external boiler evaporators operating with forced circulation.

The number of possibilities offered by the various types of evaporator is further enlarged by the choice of the boiler heating mode. Depending on the conditions and the place of operation, and depending on the capacity, it may be preferable to select a process with thermocompression or mechanical recompression of the steam produced by the evaporator, recycled as a heating fluid.

Based on this experience, SGN has developed an optimized evaporation system to treat low- and medium-level liquid radwaste commonly found on reactor sites and in nuclear research centres. The system offers outstanding decontamination performance (DF normally ranging from  $10^6$  to  $10^7$  for non-volatile species), excellent reliability, and easy operation.

A circulation pump can be used for this type of radwaste, and this has led to the proposal of forced circulation evaporation with submerged flash, offering several advantages:

- operating stability in a wide range of evaporation throughputs;
- high flow speed in the recirculation loop and the boiler, which helps to prevent the deposition of suspended matter on the heat exchange surfaces;
- boiling in the evaporation chamber where flash is developed: thus the absence of effluent evaporation and concentration in the boiler circuit prevents its scaling; and
- the "submerged flash" solution, combined with a suitable technology, helps to reduce droplet entrainment, generally very high in flash systems, to levels comparable to those found in conventional boilers.

The concentration factors obtained obviously depend on the initial salt content of the effluent and on the solubility of these salts. Yet values in the range of 10 to 100 can be obtained routinely. In the treatment of effluents containing sodium nitrate, for example, the concentrates obtained have a salt content as high as 750 g/l.

Evaporation can be performed after prior neutralization of the effluents or on acidic effluents. With acid effluents, evaporation helps to recover the acid and to avoid the storage and conditioning of substantial amounts of salts. Corrosion risks can be reduced by choosing "noble" materials, or by operating under partial vacuum at lower temperature, which significantly reduces the corrosion kinetics.

Note that the steam/liquid separation stage is obviously very important, because the distillate obtained after condensation must be as decontaminated as possible. A unit that promotes the separation of the steam and entrained droplets is an indispensable complement to the actual evaporator.

This is why SGN together with the CEA has conducted a Research and Development Program for the design, engineering, and industrial-scale development of steam purification systems, and the

means of combining these units to prevent the buildup of activity, to minimize maintenance problems, and to favor simplicity and decontamination factor stability within a broad flow range around the nominal throughput.

For steam purification, this program led to the combination of a scrubber equipped with Venturi trays, involving a new principle, and a parallel blades separator. The operating principle of the Venturi tray column is based on "liquid/gas" contact at each tray, by dispersion of the washing liquid in fine droplets into the gas to be purified. The steam flows at high speed through the mini-Venturis, replacing conventional bubble cap trays. This principle demands a very large specific surface area for contact, together with a high level of turbulence in the gas. This enhances transfers and ensures much higher efficiency, with a more compact column than in the conventional arrangement, in which the phases are placed in contact by simple bubbling of the gas through the wash liquid.

Tests conducted in identical conditions, on the same type of aerosol, with the same gas and wash liquid throughputs, showed that the decontamination factor of a Venturi tray is twice that of a bubble cap tray of the same size.

Experience has shown that, with a purification column equipped with a thermosyphon boiler, a column with three Venturi trays achieved a total decontamination factor over ten times that of a column with five bubble cap trays.

The parallel blades demister, developed by SGN for the purification of gases and vapors, runs on the principle of parallel blades settlers. In this unit, the gases entraining the liquid particles have a laminar flow at a speed of about 1 m/s between the parallel blades, which are spaced at intervals of a few mm.

90% efficiency can be obtained for 3  $\mu\text{m}$  water droplets entrained in steam at atmospheric pressure. Settling is still effective for 2  $\mu\text{m}$  particles. Side effects, such as Brownian diffusion, are also involved in the capture of smaller-diameter particles.

A first steam purification column has been installed on an evaporator of the Tricastin power plant. This type of tray has also been selected for the evaporators of the Sizewell B power plant in the United Kingdom.

Many industrial projects have resulted (design, construction, and commissioning) in response to the wide variety of client needs, either for the concentration of salt solutions (concentration of borated effluents in PWR power plants) or for purification considering the very high DF obtainable in order to meet increasingly stringent release standards.

The systems have since extended from low-capacity forced-circulation evaporators (about 250 l/h) to evaporators of the effluent treatment stations of PWR and BWR power plants, including larger-capacity evaporators (6 to 9  $\text{m}^3/\text{h}$ ) used at the Marcoule STE facility, for example, or intended for the future Japanese Rokkasho Mura reprocessing plant.

### 3. Chemical Coprecipitation of LL and ML Radwaste

In this process, the radionuclides in the effluents to be treated are made insoluble by coprecipitation, absorption and ion exchange with precipitates of inactive chemical elements, by the introduction of carefully selected reagents and the adjustment of clearly-determined pH and redox potential conditions, followed by the separation of the precipitates formed in the liquid phase.

A chemical process is hence always combined with a separation technology to produce a purified aqueous phase and a solid phase rich in radionuclides, that will have to be conditioned for disposal.

The first processes developed in the 1960s by the CEA were very similar to conventional water treatment techniques. However, these processes soon proved to be inadequate in terms of CF and DF. On completion of a number of major R&D projects conducted by the CEA teams at Cadarache, specific treatments by  $\alpha$  and  $\beta/\gamma$  emitting radionuclides were developed:

- $\alpha$  emitters

At pH  $>10.5$ ,  $\alpha$  emitters, like Pu-239 and Am-241, coprecipitate with hydroxides (especially ferric). DFs higher than 1,000 are obtained in the absence of solvent or chelant.

This decontamination can be improved by the introduction of titanium sulfate.

- Examples of  $\beta,\gamma$  emitters

- Strontium-90

Coprecipitation with barium sulfate in basic medium at pH  $>8.5$ .

DF = 50 ~ 100.

But also precipitation of:

- Ca or Fe phosphate at pH 12,
- Ca carbonate at pH 10.5 to 11, and
- Mn dioxide at pH  $>11$ .

- Cesium-137 and cesium-134

Addition of preformed nickel ferrocyanide precipitate (PPFNI) at pH 8.5.

DFs over 100 and even 1000 depend on the pH at the end of treatment.

Several radionuclides are normally present in the effluent, making it necessary to form different types of precipitate simultaneously. The optimal conditions for their formation (pH, stirring, and residence time) are unfortunately too often antagonistic, and SGN has developed industrially-commisioned units consisting of lines of stirred reactors in cascades, each operating with the best operating parameters.

This process has been used extensively by COGEMA to treat LL and ML effluents generated by its reprocessing plants, up to capacities of 100,000 m<sup>3</sup>/year for the STE3 facility (17 m<sup>3</sup>/h continuously).

SGN has accordingly benefited from many industrial references for large-capacity installations such as STEL at Marcoule and STE2 at La Hague, which has been operating continuously since its commissioning in 1966, and has processed 710,000 m<sup>3</sup> of liquid waste in accordance with the following average performance observed:

- DF total  $\beta/\gamma$  20 to 50
- DF total  $\alpha$  >1000
- DF Ru 5 to 50
- DF Cs >100
- DF Sr 50
- Concentration factors between 30 and 60.

The large volumes to be treated have led to the selection and development of continuous

processes, operated successfully thanks to the development, simultaneously with the development of the chemical processes themselves, of original technologies concerning the supply of active effluents at controlled flow rates without any mechanical equipment requiring maintenance, continuous pH and redox potential adjustments, and reagents introduction at controlled flow rates, with all these functions covered by central remote control in a nuclear environment, where the effluents contain not only  $\beta/\gamma$ , but also  $\alpha$  radionuclides.

The latest installation, STE3, was designed and built by SGN for COGEMA and has been treating LL and ML effluents from La Hague since December 1987, particularly from the UP3 and UP2 800 plants, with the STE2 facility mentioned above only used to treat "suspect" effluents.

Improvements to the chemical process applied, COGEMA's determination to optimize waste management and effluent segregation in the generating facilities, and the general policy for the new facilities of increasingly sending the effluents after in-situ concentration to the vitrification facilities, have led to treat volumes much smaller than the initial design capacity, namely, 55,000 m<sup>3</sup> of LL/ML effluents in 1991, and especially to a substantial reduction of liquid releases, for example:

- 8250 Ci  $\beta/\gamma$  in 1990 and 3130 Ci  $\beta/\gamma$  in 1991, and
- 7 Ci  $\alpha$  in 1990 and 4 Ci  $\alpha$  in 1991.

The choice of a chemical treatment process was justified by the appropriateness of these processes for the treatment of effluents containing salts, the "stable" properties obviating the need to have to make frequent adjustments in reagent types and flow rates. In addition, the rather modest overall performance ( $DF = 10^2$  to  $10^3$  and  $CF < 10^2$ ) remained compatible with the release standards in force at the time for French reprocessing plants.

For other operators besides COGEMA, this process was used for very specific applications in which the effluent properties made them unsuitable for evaporation, for example if they incurred foaming or fouling risks, or with treatment by ion exchange due to the saturation of the sites by the inactive salts present. Laundry effluents containing detergents, and decontamination effluents, are treated in this way.

In the final step, to facilitate settling and/or separation of the precipitates formed and containing the activity, it is necessary to flocculate them.

Flocculation is designed to neutralize the electrokinetic potential or zeta potential of the particles, either by the formation of hydroxides (ferric or aluminium) or by the addition of organic polyelectrolytes. Slow stirring is necessary to avoid breaking the flocs formed.

Liquid/solid separation generally takes place by static settling of the sludges followed by final clarification of the supernatants, but techniques such as centrifuging and filtration are also fully within the scope of SGN.

Note that SGN, together with the CEA, also has development programs under way on innovative techniques such as ultrafiltration, which makes it possible to achieve very high separation efficiencies for the precipitates formed, thanks to the low porosities of the membranes used, and also ensuring high concentration factors. A wide range of materials is available today for the membranes, including inorganic materials that are perfectly suitable for use in the treatment of LL and ML effluents.

# RESEARCH ON REMOVAL OF RADIOIODINE BY CHARCOAL

LI Wangchang HUANG Yuying WU Yianwei  
JIA Ming GUO Liangtian

China Institute for Radiation Protection

## ABSTRACT

The major R&D work carried out in the CIRP laboratory on removal of radioiodine is introduced, which involves the adsorption performances of various kinds of fruit shell base and coal base charcoal impregnated with chemicals, the influence of various parameters, the technique of non-destructive test for commercial scale iodine adsorber, and the iodine samplers for both gross iodine and iodine in different forms. The experimental results have been applied to the design and test of iodine adsorber and the monitoring of airborne radioiodine.

### 1. Introduction

The removal of airborne radioiodine is always one of the main subjects of air purification in a nuclear power plant for the sake of reducing the environmental radiation impact. Airborne radioiodine occurs in the form of aerosol or gas. The latter includes elemental and organic iodine. Aerosol iodine can be trapped by filters and gaseous iodine normally by charcoal. Untreated charcoal has high adsorption efficiency only for the elemental iodine. But when it is impregnated with some chemicals, the activated charcoal can effectively adsorb both organic and elemental iodine.

### 2. Study on Iodine Adsorption Performance of Impregnated Charcoal

The performance of impregnated charcoal depends mainly upon the sorts of charcoal and the composition of impregnant. Several kinds of charcoal were examined on the dynamic adsorption capacities for elemental iodine. Among them, Beijing coconut charcoal, Shanghai oil palm charcoal, and Jilin wild walnut charcoal were selected. For the impregnant selection, solutions  $KI$ ,  $KI_3$ , TEDA, and their mixtures in different concentrations were used to impregnate the wild walnut charcoal. The removal performances for methyl iodine (MeI) were tested using these impregnated charcoals. Some of the test results are given in Table 1.

In order to examine whether the selected charcoal can meet the requirements for nuclear air purification, test conditions were determined on the basis of ASTM standard D-3803 and also the needs in engineering design and the specific conditions of the laboratory. Several test results are given in Table 2. These results indicate that most of the charcoals are satisfactory in performance. For comparison, the test results of two kinds of U.S. impregnated charcoal are also listed in Table 2.

Since not only charcoal sorts but also operating conditions affect the iodine removal efficiency, the influences of several parameters were studied, including gas velocity, relative humidity (R. H.), and temperature on the MeI removal efficiency of wild walnut charcoal impregnated with 5% TEDA. The results are given in Tables 3, 4, and 5 respectively. It can be seen that the efficiencies decline with increase of gas velocity and relative humidity. Under lower relative

humidity (<40%), increase of temperature has no obvious effect on the efficiency, and under higher R. H., the increase of temperature is in favor of the adsorption of MeI.

The relationship between penetration time and concentration of MeI is emphasized in design of adsorber. Fig. 1 shows that in the range of 2-1000  $\mu\text{g/l}$  there was a linear relation between penetration time and concentration on log-log plot. The minimum MeI concentration used in the test was 2  $\mu\text{g/l}$ , higher than the actual concentration in nuclear power plant by a factor of  $10^6$ . In such a consideration, penetration time will probably be much longer than the figure shown in Fig. 1.

The test results mentioned above have been used as the basis of the design of iodine adsorber Dx 1700 I with 2.5 cm thick charcoal. This kind of adsorber has been adopted in some nuclear facilities.

In some countries, there appeared the tendency that the fruit shell charcoal used in iodine adsorber is replaced by coal base charcoal and the thin bed adsorber, by deep one. Some research work has been carried out in the CIRP laboratory.

Although fruit shell charcoal has higher adsorption capability and mechanical strength, they are very expensive because of the limited sources. In recent years, production of high-quality coal-base charcoal has been developing rapidly in China. With some domestic coal-base charcoal impregnated with 5% TEDA, the performance test has been conducted to study the possibility of applying it in nuclear air purification. The test results are given in Table 6. Additionally, the impregnated Dx-15 charcoal has been tested according to ASTM D3803, the results of test A, B, and C are listed in Table 7. As compared with Table 1, the coal-base charcoal is comparable to fruit shell charcoal in performance. So it can be used in iodine adsorber as a substitute for fruit shell charcoal.

In practice, it has been noted that the service life of thin layer adsorber is often shortened due to aging and poisoning of charcoal. Poisoning is caused by organic vapor occupying the active sites of charcoal, whereas aging is the results of long period exposure of charcoal to ambient air. To study the effects of the two processes, artificially aged and poisoned charcoal has been compared with unused one on their adsorption performance. The results are shown in Fig. 2. In semilog plot, the MeI penetrations through new charcoal bed decrease sharply with increase of bed thickness, and those through aged and poisoned charcoal bed decrease slowly in the front section but quickly in the rear section with increase of bed thickness. This shape of penetration curve shows that the front section serves as guard bed so that the rear section can maintain its normal adsorption capability. This reflects the advantages of deep bed adsorbers.

### 3. Non-destructive Leak Test for Iodine Adsorbers

The efficiency of iodine adsorber depends upon not only the charcoal properties but also its manufacture quality. As a result of improper design or manufacture, or imperfect filling of charcoal layer (uncompacted, nonuniform), channels would occur in charcoal, which will cause leakage and decrease of absorption efficiency. To ensure high efficiency of the adsorber, non-destructive leak test must be carried out before it is used.

In this test, Freon 112 (F-112) is used as a tracer. By comparing the penetration rate of F-112 through adsorber, the quality of the manufactured adsorber can be identified. In case of an

adsorber with satisfactory performance, the penetration rate shall be less than 0.01% when the time extrapolated from the penetration curve is 0.

In a non-destructive test, the following conditions should be met:

- (1) The upstream F-112 concentration shall be no less than  $10^4$  times the detection limit of the instrument; and
- (2) The load of F-112 on charcoal in each test shall be as small as possible so that the test can be carried out repeatedly.

On the basis of laboratory study, the test conditions are selected as follows:

Upstream F-112 concentration  $20 \times 10^{-6}$  (v/v)

Gas velocity  $> 1.5 \text{ l/min} \cdot \text{cm}^2$

Temperature of gas stream  $< 40^\circ\text{C}$

Relative humidity of gas stream  $< 70\%$

Water content in charcoal  $< 37\%$ .

The leak test equipment for commercial scale adsorber was designed and installed, as shown in Fig. 4. The equipment consists of test system, F-112 generator, and sampling system. To get the representative samples, multipoint sampler was designed. With this equipment, following operation conditions could be obtained:

Gas flow rate:  $500 \sim 2100 \text{ m}^3/\text{h}$

Gas temperature: ambient temperature to  $50^\circ\text{C}$

F-112 generation rate:  $0.25 \sim 50 \text{ g/min}$

Pressure drop:  $2.2 \text{ kPa}$  (when flow rate is  $1700 \text{ m}^3/\text{h}$ ).

The test for one set of absorber can be completed in 5 minutes. This equipment is used for routine delivery inspection.

#### 4. Development of Radioactive Iodine Sampler

In routine or emergency radiation monitoring, radioiodine is one of an important items. In order to obtain the representative samples of gaseous radioiodine, two kinds of iodine samplers have been developed in the laboratory: one is sampler DQ-01 for gross iodine; the other is for iodine in different forms.

The sampler DQ-01 shown in Fig. 4 consists of several  $\phi 50 \text{ mm}$  polyethylene cells filled with 5% TEDA impregnated coconut shell charcoal. The sampling efficiencies of cells with different thickness were examined under various gas velocities ( $100 \sim 200 \text{ l/min}$ ), temperatures ( $13 \sim 40^\circ\text{C}$ ) and relative humidities (56~95%). Even under severe conditions, e.g.  $200 \text{ l/min}$  gas velocity,  $35^\circ\text{C}$ , and 94% R.H., the efficiency of the sampler with 2 cm thick charcoal is near 100% for elemental iodine in 2 hours of sampling. For MeI, with 6 cm thick charcoal, the efficiency is over 97%. The thickness of charcoal bed and air flow rate can be adjusted according to the environment conditions.

Since different forms of iodine have different radiation effects, and different physical and chemical properties, it is necessary to determine the concentration of various forms of iodine to correctly assess radiation effects and to select proper cleaning process. For this purpose, a kind of sampler has been developed. Polytetrafluoroethylene has been used as the material of sampler casing in order to minimize the iodine adsorption on the wall. The sampler is composed of

several sections which are connected one another by screws and sealed with O rings (see Fig. 6). For this sampler, suitable materials are chosen to trap iodine selectively, фп-25 high efficiency glass fiber paper (material 1) for aerosol iodine, carrier 6201 impregnated with CdI<sub>2</sub> (material 2) for elemental iodine, and coconut shell charcoal impregnated with 5% TEDA (material 3) for organic iodine. The efficiencies of various trapping materials have been examined under various conditions involving bed thickness, gas velocity, temperature, and relative humidity, and the performance of the combination of these materials are also tested. The test results are given in Table 8. This kind of sampler has been used on site, the results are shown in Tables 9 and 10.

Table 1 Methyl Iodide Removal Efficiency (%) of Different Impregnated Charcoal

Charcoal section	Content of impregnant				Charcoal W. W. -201
	4% KI <sub>3</sub>	3% KI	5% TEDA	2% KI+4% TEDA	
0-1	82.50	81.31	95.0	94.67	49.72
1-2	14.52	14.89	4.80	5.06	29.73
2-3	2.40	3.32	0.18	0.24	13.05
3-4	0.42	0.39	0.02	0.03	5.23
4-5	0.10	0.04	0	0	1.74
5-6	0.06	0.04	0	0	0.43
Charcoal particle size	8—20 mesh				
Gas velocity	15 m/min				
Concentration of methyl iodide	32 mg/l				
R. H.	40%				
Temperature	40°C				
Test duration	2 h				

Table 2 Test Results for Six Impregnated Charcoals

Charcoal *	Penetration (%)					Efficiency (%)
	Test 1	Test 2	Test 3	Test 4	Test 5	
Charcoal 1 + 5%TEDA	0.05	0.2	0	0.02	99.97	
Charcoal 1 + 4%KI <sub>3</sub>	0.2	0.2	0.3	0.06	99.98	
Charcoal 2 + 5%TEDA	0.05	0.04	0.8	0.01	100	
Charcoal 2 + 4%KI <sub>3</sub>	0.7	0.16	0.8	0.01	99.92	
Charcoal 3 + 5%TEDA	0.02	0.04	0.03	0.04	99.90	
Charcoal 3 + 4%KI <sub>3</sub>	1.7	0.4	0.2	0.04	99.84	
U. S. Bc-727	4.2	0.8	0.34	0.15	99.50	
U. S. Nusorb KIETG 2	6.2	7.6	0.02	0.00	99.99	
Acceptable value	<3	<1	<2	<0.1	<99.5	

\* Charcoal 1—Beijing coconut shell charcoal

Charcoal 2—Wild walnut charcoal-201

Charcoal 3—Oil palm charcoal-1

Table 3 Methyl Iodide Removal Efficiencies (%)

Charcoal section	Gas velocities 1/min · cm <sup>2</sup>			
	1.0	1.5	2.0	2.5
0-1	98.55	96.60	95.40	89.52
1-2	1.40	3.23	4.46	9.82
2-3	0.05	0.07	0.14	0.63
3-4	0	0.10	0	0.03

Table 4 Methyl Iodide Removal Efficiencies (%)  
versus Relative Humidities

Charcoal sections	Relative humidities (%)			
	40	60	80	95
0-1	96.61	91.67	83.60	72.58
1-2	3.32	8.15	14.95	22.58
2-3	0.07	0.18	1.45	4.01
3-4	0	0	0	0.83
4-5	0	0	0	0

Table 5 Methyl Iodide Removal Efficiencies versus Temperatures

Charcoal sections	Temperatures (°C)					
	20°	40°	60°	80°	40° °	80° °
0-1	97.30	96.60	96.90	96.30	72.58	86.78
1-2	2.60	3.30	3.90	3.61	22.58	11.68
2-3	0.10	0.10	0.10	0.09	4.01	1.35
3-4					0.83	0.19

\* R. H. = 40%

\*\* R. H. = 95%

Table 6 Methyl Iodide Removal Efficiencies (%)  
of Impregnated Coal-base Charcoals

Charcoal	Thickness of charcoal bed (cm)			
	1	2	3	4
DH-30+5%TEDA	96.66	99.34	99.90	100
DZ-30+5%TEDA	95.44	99.16	99.74	100
Dx-30+5%TEDA	95.34	99.52	99.97	100
Dx-15+5%TEDA	94.47	99.36	99.87	100

Table 7 Test Results for Impregnated Coal-base Charcoal

Test	Methyl iodide penetration (%)		
	Test A 30°C, R. H. 95%	Test B 80°C, R. H. 95%	Test C 130°C, R. H. *
	Acceptable value	3	1
Test result	0	0.15	0

\* Saturated steam at 100°C is heated up to 130°C.

Table 8 Adsorption Efficiencies of Combined Sampler for Elemental Iodine and Methyl Iodide

Glass fibre paper			CdI <sub>2</sub>			Charcoal+TEDA			Charcoal+TEDA		
1*	2	3	1	2	3	1	2	3	1	2	3
1.84**	1.11	0.88	95.13	0.60	0.11	0.20	0.13	0	0	0	0
0.11***	0.06	0.08	0.02	0.02	0.005	97.26	2.37	0.05	0	0	0

\* layer of adsorbent

\*\* for elemental iodine

\*\*\* for methyl iodide

Table 9 Distribution of Elemental Iodine-131 in the Sampler (%)

Test No.	Iodine concentration $10^{-2}$ Bq/l	$\phi\pi\pi-25-1$		Carrier 6201+CdI <sub>2</sub>	
		Layers 1+2	Layer 1	Layer 2	Layer 3
P 1-1	82.7	0	98.12	0.81	1.12
P 1-2	1128	0.07	99.78	0.13	0.02
P 1-3	268.3	1.24	86.29	11.49	0.98
average		0.44	94.70	4.14	0.71

Table 10 Distribution of Organic Iodide on Charcoal (%)

Test No.	Concentration of organic iodine $10^{-2}$ Bq/l	Charcoal layer		
		1	2	3
P 1-2	9.0	97.94	2.18	0
P 1-3	16.8	90.07	5.96	3.97
Average		94.01	4.07	1.99

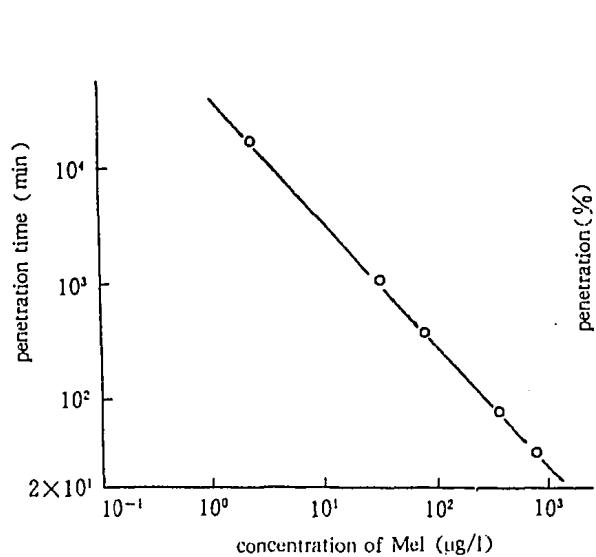


Fig. 1 Effect of concentration on penetration time

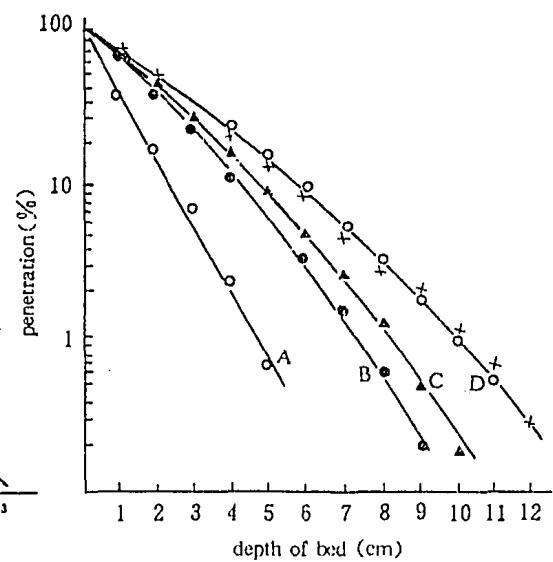
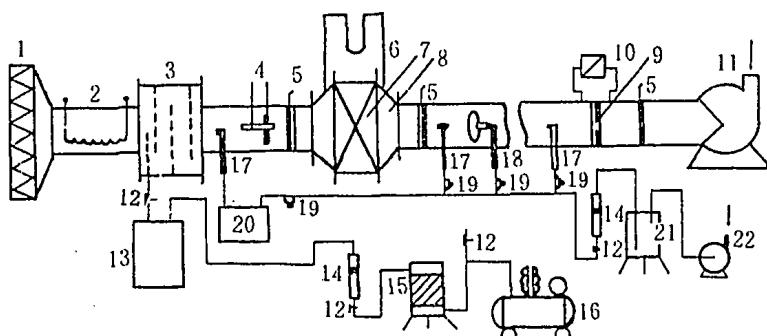
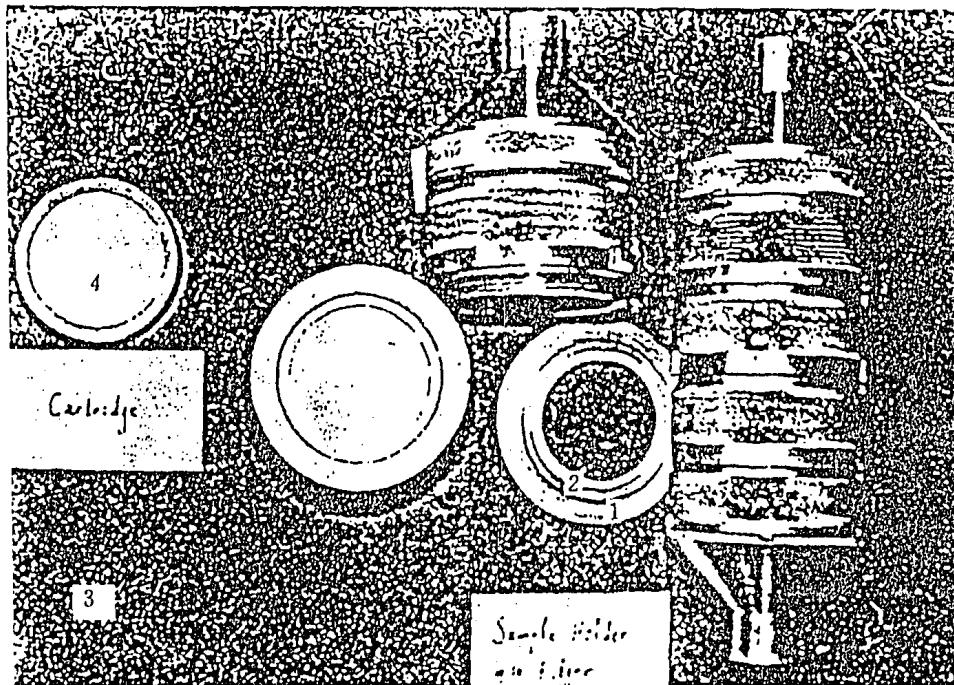


Fig. 2 Effect of aging and poisoning



- |                       |   |                                |
|-----------------------|---|--------------------------------|
| 1. HEPA filters       | 9. Flowmeter                            | 16. Compressor                 |
| 2. Heater             | 10. Declination type differential gauge | 17. Single point sampler       |
| 3. Gas mixer          | 11. Fan                                 | 18. Multi-point sampler        |
| 4. Hydroscope         | 12. Adjusting value                     | 19. Sampling port              |
| 5. Sealing valve      | 13. F-112 vapor generator               | 20. Diluting system of samples |
| 6. Differential gauge | 14. Rotating flowmeter                  | 21. Buffer                     |
| 7. Iodine adsorber    | 15. Oil trap                            | 22. Vacuum pump                |
| 8. Clamping device    |   |                                |

Fig. 3 Leak test equipment



1. Holder                            2. Press ring of HEPA paper  
3. HEPA filter paper                4. Cartridge

Fig. 4 Gross iodine sampler

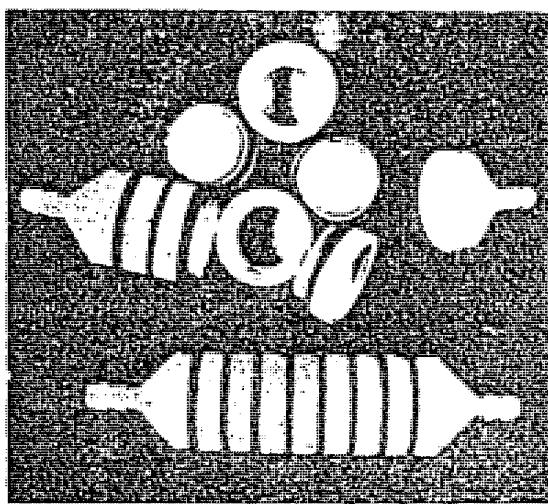


Fig. 5 Sampler for iodine in different forms

# VOLUME REDUCTION OF LOW- AND MEDIUM-LEVEL WASTE BY INCINERATION/CALCINATION

A. de BUZONNIERE J. C. GAUTHEY  
TECHNICATOME, FRANCE

## 1. Introduction

Nuclear installations generate large quantities of low- and medium-level radioactive waste. This waste comes from the various installations in the fuel cycle, reactor operation, research institutes, hospitals, nuclear plant dismantling, etc.

The low- and medium-level waste is mainly technological; solvents and oils, overshoes, gloves, slurries, etc.

Of the waste treatment processes, incineration has the following advantages:

- considerable reduction in the initial volume of waste and
- mineralization and stabilization of waste, enhancing the safety of final storage.

For a number of years, TECHNICATOME has been examining, developing, and producing incineration and drying/calcination installations. These include:

- a waste oils and solvents incinerator,
- a solid and liquid waste incinerator, and
- slurries dryer/calciner.

These three installations are described below.

## 2. Liquid Waste Incinerator

Technicatome did the project development work for the incineration plant of PIERRELATTE (France) on behalf of COGEMA. This plant has been in active service since November 1987.

### 2. 1 Waste characteristics

The following waste is incinerated:

- oils contaminated with U-238 and U-235,
- solvents contaminated with uranium, and
- scintillation liquids.

The physical, chemical, and radiological characteristics are given in Appendix 1.

### 2. 2 Installation capacity

This corresponds to a load of 525 thermies/hour; either 75 kg/h of oils with an NCV of 30,000 kJ/kg (7,000 kcal/kg), or 50 kg/h of solvents with an NCV of 44,000 kJ/kg (10,500 kcal/kg).

The volume of liquid waste burned to date is about 1,000 m<sup>3</sup>(80% oils and 20% solvents).

### 2. 3 Design of the unit

Among the design criteria selected by TECHNICATOME such as radioactivity, containment,

waste volume reduction, and risks specific to the waste to be incinerated are taken into account, namely :

- previous contamination of oils by the product resulting from the decomposition of uranium hexafluoride (this entailing criticality enriched Uranium) and corrosion (possible formation of hydrofluoric acid), and
- risk of fine outbreaks and explosion of various solvents.

The process diagram is given in Appendix 2.

#### **2. 3. 1 Waste storage**

The liquids to be burned are stored in two tanks:

- a 10 m<sup>3</sup> oil tank agitated by a mixing loop and fitted with an external coil steam heater and
- a 10 m<sup>3</sup> solvent tank fitted with a mechanical agitator and a nitrogen neutralizer.

A 500 litre oil tank located in the building enables the right temperature to be maintained in the quantity of oil needed to startup the boiler.

Preparation of the solvents is a delicate process in order to prevent gelling, and involves successive filtering operations.

#### **2. 3. 2 Incineration**

The incinerator is a fixed horizontal boiler with smoke tubes equipped with an oil/solvent dual fuel burner.

The solvents are injected in the burner through an injection rod.

The furnace outlet temperature is 900°C to 1,000°C.

The combustion gases are cooled in the smoke tubes to 250~300°C.

#### **2. 3. 3 Primary filtration**

Slight dilution with ambient air allows the negative pressure in the gas circuit to be adjusted.

A multicyclone traps dust from the gases before washing.

#### **2. 3. 4 Gas washing**

The gases are washed by a two-stage venturi with a soda solution in order to:

- neutralize at 60°C the acids formed during combustion and
- supplement dust extraction from the smoke.

The gas washing waters are periodically changed and sent to the waste processing station.

#### **2. 3. 5 Final filtration**

On leaving the washing plant, the gases undergo final filtration consisting of:

- a demister for extraction of droplets from the gases,
- heating to about 80°C with an electric battery in order to limit the risks of condensation during extraction, and
- filtration of radioactive particles on the THE filters.

### **2. 3. 6 Extraction**

The gases are extracted to the stack by an extraction fan which maintains a negative pressure in the installation.

An isokinetic sampling device is used to check the gases released to the stack (by measuring the activity deposited on a filter).

### **2. 3. 7 Evacuation of ashes**

The ashes are recovered in the containment:

- in soot traps (by suction device),
- in the drum placed under the multi-cyclone, and
- in the washing waters, by filtration through a cartridge filter.

The ashes are then sent to the waste processing station where they are drummed.

### **2. 3. 8 Operation**

The installation is operated in 100-hour runs (from Monday morning to Friday afternoon) by teams of 2 operators.

During shutdowns, the unit is maintained and the next batch of liquid waste is prepared (with analysis and uranium balance).

The average production of ashes is 5 to 10 kg of ash per m<sup>3</sup> of liquid waste burned.

The proportion of unburned matter in the ashes is 2%.

The regulated operating parameters are as follows:

- the oil heating temperature at the incinerator injection rod inlet,
- the negative pressure in the combustion gases circuit,
- the filling level of the oil preparation interim tank, and
- the nitrogen pressure in the roof of the solvents tank.

An hourly monitoring sheet is filled in to check the evolution of the various operating parameters (levels, flowrates, temperatures, and differential pressures).

### **2. 3. 9 General layout of the installation**

The incinerator and associated gas treatment plant are installed in a building 13.5m×8m×7m. The discharge areas and storage tanks are installed in a hangar adjoining the building and separated from it by a fire-wall and a retention pit (14m×6m×6m).

The unit is controlled from a separate control room so as to limit the risks of explosion and fire.

## **3. Solid and Liquid Waste Incinerator**

Technicatome was chosen by BATAN for designing and building Radwaste Management Stations for the National Nuclear Research Center. In addition, Technicatome was in charge of the Incinerator by a turnkey contract. This incinerator was commissioned in 1992. Appropriate technical solutions were chosen considering the customer's needs and Technicatome's experience through operational installations existing in France.

### **3.1 Waste characteristics**

The original feature of this incinerator is its ability to burn liquid, solid, and biological waste. The waste incinerated is:

- solid PVC, cotton and polythene based technological waste,
- biological waste: small animal corpses, and
- liquid waste.
  - lubrication and drainage oils,
  - scintillation liquids, and
  - tributyl phosphate diluted in dodecane.

The radiological characteristics are given in Appendix 3.

### **3.2 Installation capacity**

The incineration capacity is 50 kg/h for solid waste with an NCV of 5000 kcal/kg, or 20 kg/h for liquid waste with an NCV of 10,000 kcal/kg.

### **3.3 Description of the installation**

The process diagram is given in Appendix 1.

#### **3.3.1 Waste storage and preparation**

- Solid technological waste:

Solid technological waste is handled in a glovebox for separation of combustible from non-combustible waste. The combustible waste is placed in plastic bags, themselves placed in cardboard boxes.

- Biological waste:

Tight plastic bags of biological waste are stored in freezers.

- Liquid waste:

Liquid waste containing TBP is stored in a 100 litre tank. Liquid waste not containing TBP is stored in a 1000 litre tank. The liquid waste is homogenized in a 1500 litre tank before being injected into the incinerator. The liquids containing TBP are first mixed with the previous liquids to ensure a TBP concentration of less than 3% in the final mixture.

#### **3.3.2 Incineration**

The incinerator is of the controlled air type. It comprises 2 vertical chambers;

- the 1st chamber: low-oxygen combustion for solid waste, loaded in 5 to 7 kg batches (temperature between 500 and 800°C) and
- the 2nd chamber: high-oxygen post-combustion for gases from the 1st chamber during solid waste combustion. The temperature in this chamber is between 1,000 and 1,200°C.

The burners in each of the chambers are equipped with three settings for adapting the heating power to the heat energy required by waste combustion.

The solid waste boxes are loaded into the incinerator on a sloping conveyor. They then enter the furnace through a double air-lock.

The liquid waste is injected into the 2nd chamber of the incinerator by a metering pump from a storage tank located in an adjacent sunken pit located outside the building. This tank is fed with

nitrogen.

In the case of incineration of liquid waste containing TBP, the phosphoric acid created during combustion is neutralized inside the furnace.

### **3. 3. 3 Dilution**

On leaving post-combustion, the gases are cooled by dilution with air to lower their temperature to 180°C in a third chamber above the two combustion chambers. The air supply comes from a variable speed fan.

### **3. 3. 4 Filtration**

After dilution, the gases are filtered through two stages at 180°C:

- filtration in a bag filter with automatic continuous compressed air unclogging and
- filtration on THE filters.

### **3. 3. 5 Gas washing**

Gas washing with a soda solution circulating in a venturi washer ensures:

- cooling of the gases to about 50°C and
- neutralization of the acids formed during combustion.

The gases thus neutralized are then dehumidified in a lined column and then in a demister leaving the column.

The washing water is periodically sent to the waste processing station for control before release.

### **3. 3. 6 Gas extraction**

Before extraction to the stack, the gases are heated to 60°C by an electric heater in order to limit the risk of condensation during extraction.

The gases are extracted by a variable-speed fan, which also maintains the negative pressure in the circuit.

A continuous check is run on the activity of the gases prior to release to the atmosphere.

### **3. 3. 7 Evacuation of ashes**

The ashes are extracted from the 1st combustion chamber and from the foot of the bag filter via an airlock in which they are cooled before being recovered and confined in a metal drum.

The ashes are then encapsulated in cement with mixing by a drum-turner.

### **3. 3. 8 Operation**

Waste is incinerated in 6-day runs. Each day comprises a period of 6 hours during which the unit incinerates waste and a period of 18 hours during which the unit is kept at temperature (thermal standby).

Ashes are extracted from the furnace daily before the incineration period.

The unit is run from a control room separate from the incineration plant and equipped with a control panel with mimic diagram, indicators, and recorders.

The following parameters are controlled:

- the negative pressure in the incinerator by means of the extraction fan,
- the temperature in each of the 2 combustion chambers by adjusting the burner rate,
- the gas cooling temperature by means of the dilution fan, and
- the oxygen rate in the smoke by means of the combustion air valve.

Discharge of liquid waste is carried out and controlled locally.

### **3. 3. 9 General layout of the installation**

The incinerator and the waste sorting station are installed in one room.

Liquid waste storage is in a separate room.

The unit is controlled from a control room, and separated from it so as to limit the risks of explosion and fire.

The health-physics measurements are also indicated in this room.

## **4. Drying/Calcination**

The process chosen by Technicatome is the LEFLASH method developed by RHONE POULENC RECHERCHE. It has proved effective at full scale on a large number of branches in industry.

The process uses a special flow system. By means of suction in the stream of hot gases the system performs thorough splitting of the material to be dried.

### **4. 1 Waste characteristics**

The dryer-calciner was developed for the chemical and foodstuffs industry.

Depending on the proportion of dry matter in solution, it can be used in the nuclear industry:

- either as a concentrator of liquid waste with low concentration of dry matter,
- or as a calciner of liquid waste containing a low percentage of dry matter,
- or as a dryer of aqueous solutions or wet solids.

### **4. 2 Installation capacity**

The following performance figures were obtained on the pilot plant with the same drying head.

#### Concentrator

Processing of decontamination waste containing 5% dry matter (by weight) :

- processing capacity  $1.2 \text{ m}^3/\text{h}$  at 5% dry matter and
- production of 300 l/h of concentrate at 18% dry matter.

The corresponding air flow is  $1,500 \text{ Nm}^3/\text{h}$  at 0.5 bar with a consumption of 60 kg/h of gas (for instance methane).

#### Calciner

processing of previous waste containing 18% dry matter :

- processing capacity 300 l/h and
- production of a calcinated powder carried in a hot gas, separated by a bag filter.

The corresponding air flow is  $1,500 \text{ Nm}^3/\text{h}$  at 0.5 bar with a consumption of 75 kg/h of gas (for instance methane).

#### **4. 3 Installation description**

##### **4. 3. 1 Waste drying**

The product to be processed is mixed in the mixing head (Fig. 1) with air raised to a high temperature (800~900 °C for drying and 1,200 °C for calcination).

The hot gases arrive at high speed in the periphery of the "well-vortex" system.

The product to be processed is then sucked in by the negative pressure created in the axis of the vortex, and simply mechanically dispersed into the hot gases. This system allows instantaneous "flash" processing.

After the flash effect, the gases are recovered at the following temperatures:

- 800 °C in the case of the calciner,
- 120~150 °C in the case of the dryer, and
- 70 °C in the case of the concentrator.

##### **4. 3. 2 Gas processing**

When the calciner is in operation, the gases are diluted with air down to about 150 °C before filtration in a bag filter followed by final THE filtration (see Appendix 6).

When the dryer is in operation, the gases are filtered on a bag filter before THE filtration.

When the concentrator is in operation, the gases are purified in a cyclone before final filtration on a THE filter, or humid safety filtration (see Appendix 5).

##### **4. 3. 3 Extraction**

The purified gases are extracted by a fan.

##### **4. 3. 4 Evacuation of ashes**

The concentrates retrieved from the base of the cyclone, or the powders recovered from the base of the bag filter are sent away for drumming.

##### **4. 3. 5 Operation**

The operation of the ISOFLASH dryer can be adjusted for a given air flow:

- either for the "calciner" with protected walls (high gas flow, post-flash temperature higher than 800 °C for example),
- or for the "dryer" with heat-insulated walls (average gas rate leading to total evaporation of the water),
- or for the "concentrator" with bare walls (low gas rate leading to partial evaporation of the water at a post-flash temperature lower than the dew point of the mixture obtained).

##### **4. 3. 6 Advantages of the process**

The advantages of the process are as follows:

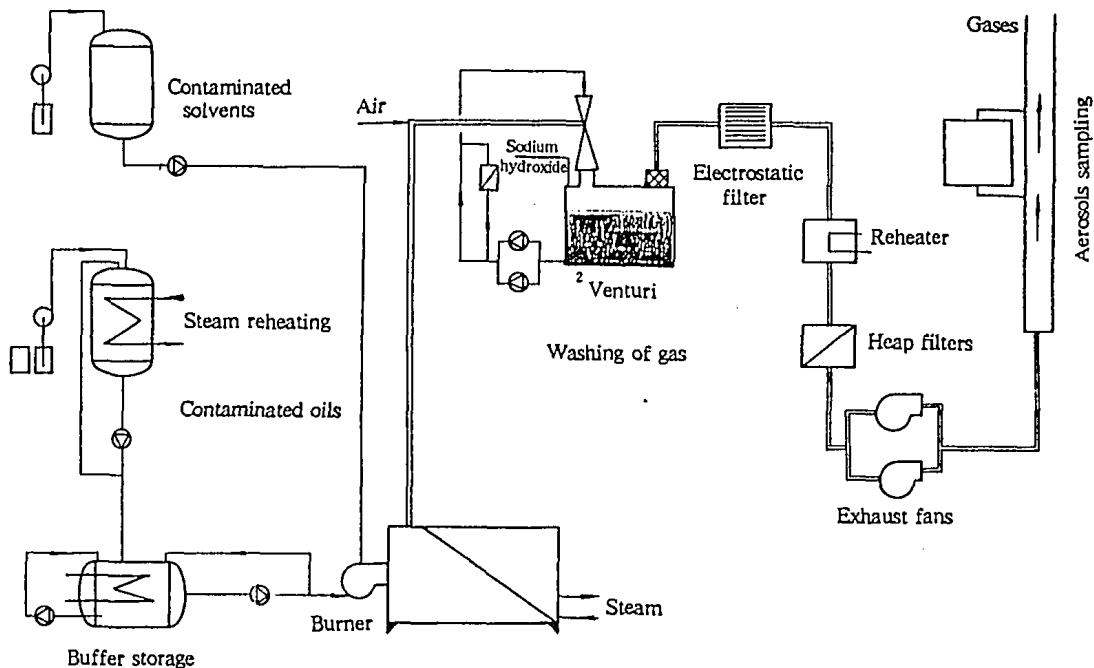
- simply designed equipment whose "drying head" is the same, whatever the type of application,
- highly compact units,
- total absence of moving mechanical parts in direct contact with the products,
- easy maintenance,
- low thermal inertia, and

- ultra-rapid, systematic, uniform calcination restoring any original granulometry, or giving a fine regular granulometry (about 2 to 10 microns).

#### Appendix 1 Liquid Waste Incinerator—Waste Characteristics

Type of waste	Physico-Chemical Characteristics	Activity
oils	<ul style="list-style-type: none"> <li>uranium content : 0.5 g/l average (5 g/l max)</li> <li>suspension matter : 2 g/l average (8 g/l max)</li> <li>chlorides : 1 g/l average (3 g/l max)</li> <li>fluorides : 1 g/l average (3 g/l max)</li> <li>water content : 15% max</li> </ul>	U-238 and U-235 contamination (0.5 g/l) Activity: $2 \times 10^4$ Bq/l and $\alpha$ in $\beta$
solvents	<ul style="list-style-type: none"> <li>uranium: 0.15 g/l (1 g/l max)</li> <li>chlorides and fluorides: 3 g/l max</li> <li>water content: 5 à 10%</li> </ul>	Contamination and uranium (0.15 g/l) Activity $< 10^4$ Bq/l and $\alpha$ in $\beta$
scintillation liquids	Solvents containing essentially toluene, cumene et xylems. Presence of plastic residue from shredding of polyethylene bottles.	Activity $< 10^6$ Bq/l

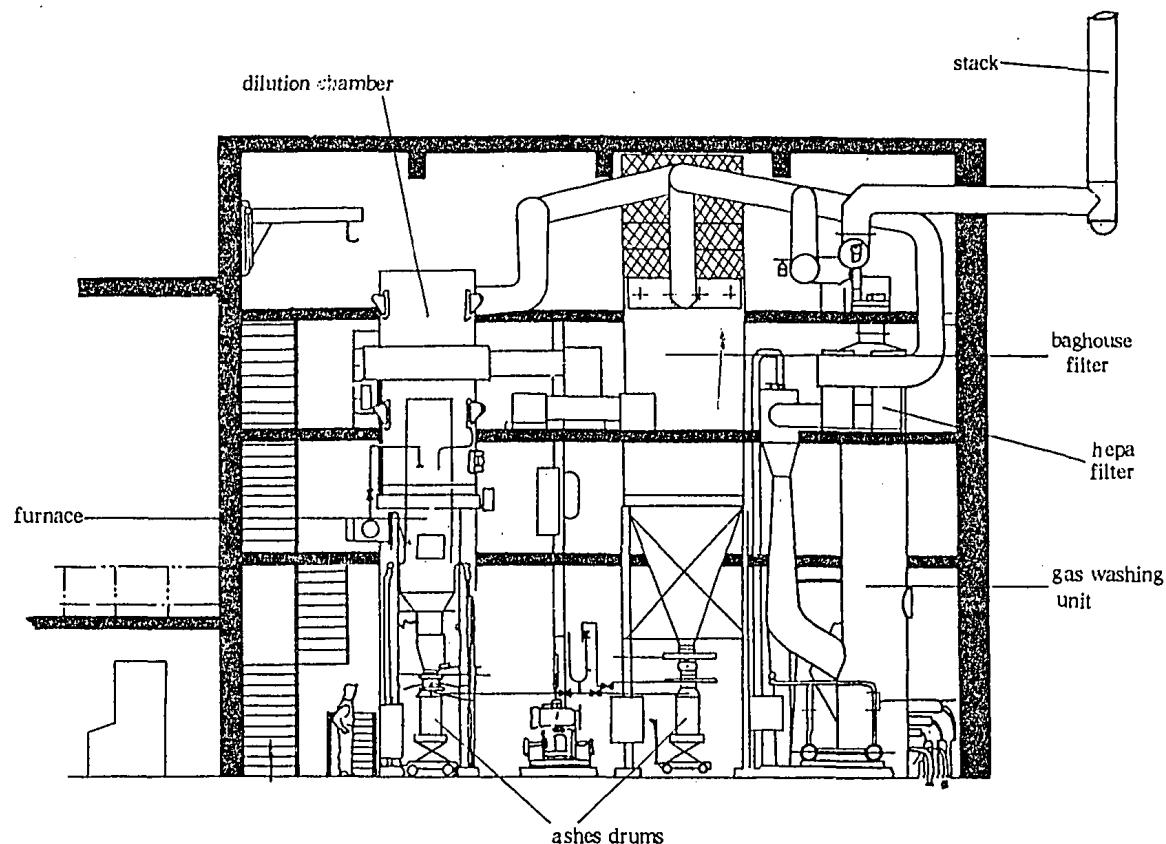
#### Appendix 2 Liquid Waste Incinerator Pirrelatte (COGEMA)



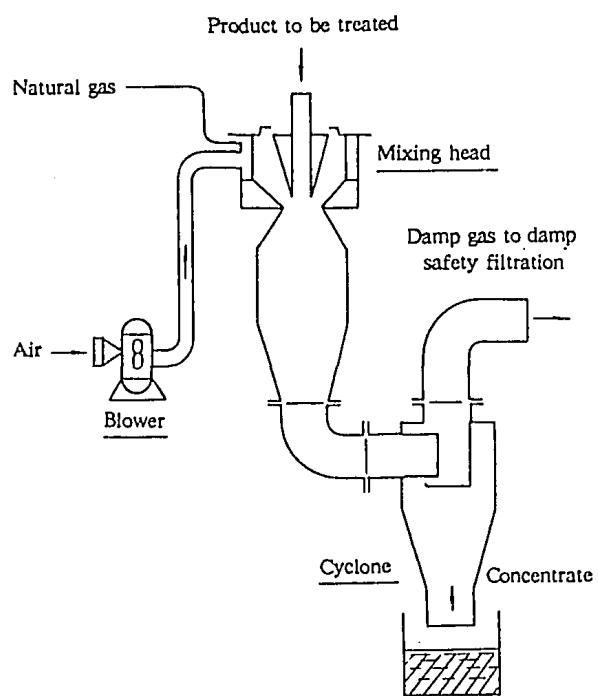
### Appendix 3 Solid and Liquid Waste Incinerator

Type of waste	Composition	Activity
solid	• Rubber polyethylene PVC, cotton, paper, cardboard, wood	$\alpha < 3.7 \times 10^6 \text{ Bq/m}^3 (10^{-4} \text{ Ci/m}^3)$ $\beta, \gamma < 3.7 \times 10^8 \text{ Bq/m}^3 (10^{-2} \text{ Ci/m}^3)$
liquid	• Drainage and greasing oils : viscosity $< 6 \text{ cSt at } 40^\circ\text{C}$ • Scintillation liquids (xylene-Toluene) steam pressure $< 130 \text{ mbar at } 30^\circ\text{C}$ • Tributyl phosphate (TBP) diluted at 20% maximum in dodecane	$\alpha < 3.7 \times 10^6 \text{ Bq/m}^3 (10^{-4} \text{ Ci/m}^3)$ $\beta, \gamma < 3.7 \times 10^8 \text{ Bq/m}^3 (10^{-2} \text{ Ci/m}^3)$
biological	Carcasses of small animals	$\alpha < 3.7 \times 10^3 \text{ Bq/kg (10}^{-7}\text{ Ci/kg)}$ $\beta, \gamma < 3.7 \times 10^5 \text{ Bq/kg (10}^{-5}\text{ Ci/kg)}$

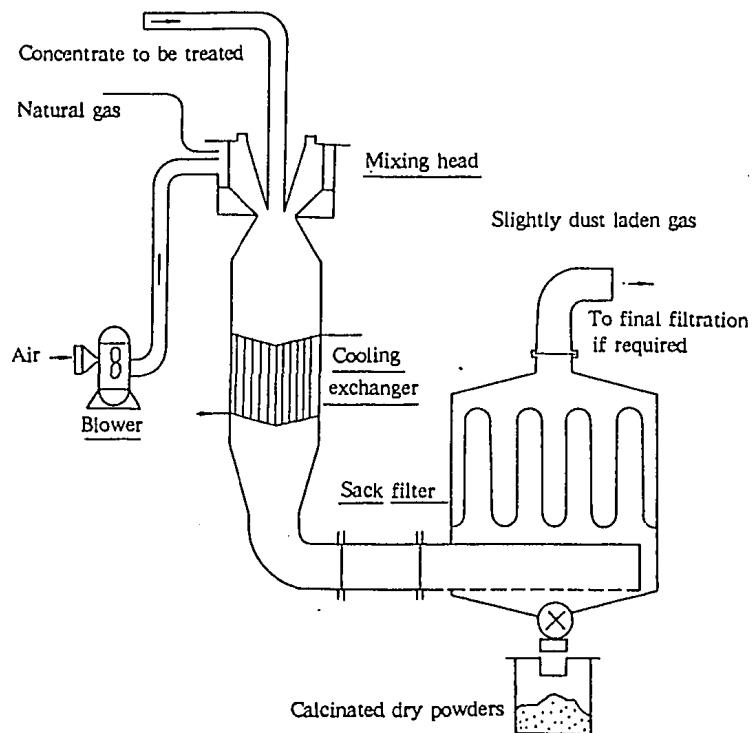
### Appendix 4 Incineration Unit for Solid and Liquid Waste



## Appendix 5 Overall Diagram of Concentrator Skeleton Diagram



## Appendix 6 Overall Diagram of Calcinator Skeleton Diagram



# INCINERATION IN THE NUCLEAR FIELD

## THE SGN EXPERIENCE

Serge CARPENTIER  
SGN, FRANCE

### 1. Introduction

The operation of power reactors, like that of fuel fabrication and nuclear fuel reprocessing plants, generates substantial quantities of waste. A large share of this waste is low- and medium-level waste, which is also combustible.

Similarly, a number of institutes, laboratories, and hospitals, in the course of their activities, generate waste of which a portion is radioactive and combustible.

It is the duty of the States that generate these wastes to manage their subsequent treatment and to ensure that they never pose a hazard to the environment.

To do so, adequate treatment is indispensable to reduce their volume for economic reasons (lower overall disposal cost) and also practical ones (to avoid quickly saturating the disposal sites).

Moreover, the treatment of these technological wastes can help to isolate hazardous substances (such as plutonium) and thus to make disposal safer.

### 2. Treatment by Incineration

The chief advantage of incineration is to minimize the volume of burnable waste treated, and to produce a residue termed "ash". This ash is usually stable from the physicochemical standpoint, as the chelatants and a large part of the organic components have been destroyed by the action of heat.

Radiowaste incineration is essentially designed to achieve:

- complete combustion with the best weight and volume reduction factor,
- fume purification, by dry or wet processes, generating a minimum of secondary waste,
- uncomplicated and hence lower-cost equipment that can be fully automated,
- installation running under constant negative pressure, offering the fullest guarantee against overpressure of explosion, and eliminating outside contamination hazards,
- long service life of refractory materials,
- acceptance of a wide range of sundry waste including oils and solvents, and
- easy evacuation of inert ash to the immobilization facility.

### 3. Incinerable Wastes

Two main classes of incinerable wastes can be distinguished according to their origin:

- $\beta/\gamma$  waste generally produced by reactors and research centers, and
- waste containing  $\alpha$  emitters produced in fuel cycle facilities, especially fuel fabrication and

reprocessing plants.

Whereas, they may differ in their type of radioactivity, incinerable wastes are basically divided by their physical character into three families—solid, liquid, and gas.

The highly-diversified solids family includes:

- plastics such as polyethylene, polystyrene, PVC, neoprene, polyurethane, etc. ,
- rubbers and elastomers,
- cellulose materials such as paper, board, rags, cotton, wood, etc. , and
- incombustible materials: metals and glass.

Solid wastes are generally mixtures of the above components. Their average low heating value ranges from 12,000 to 30,000 kJ/kg depending on the composition of the mixture and the moisture content.

The bulk density is low (about 150 to 200 kg/m<sup>3</sup>) rising to 300 to 400 kg/m<sup>3</sup> when coarsely crushed.'

The liquids family may include oils or high water content tritiated solvents (up to 30% water or more).

The oils have a specific gravity of about 0.9, viscosities at 20°C of about 50 CP, and a flash point of around 60°C. They are miscellaneous oils including cutting oils.

The gas family includes butenes and butanols from pyrolysis operations on solvents such as tributylphosphate and dodecane used in reprocessing plants.

#### 4. Incinerators — the Available Technologies

A wide variety of incinerators is available throughout the world. SGN has built up 25 years of experience in this field and has many completed projects to its credit. It is involved today in major projects (including the "alpha" incinerator of the Melox plant, the "beta/gamma" incinerator of the El Cabril center in Spain, and the centralized incinerator project in southern France) and specialized in the following processes.

- $\beta/\gamma$  incinerator: development of a standard excess air incinerator for capacities from 10 to 150 kg/h,

For higher capacities, exceeding 1 t/h, SGN is currently designing a larger incinerator with mechanized grates and a heat boiler installed on the fumes circuit.

Concerning oils and solvents, SGN is fitting these incinerators with a high efficiency combustion system, specially designed at the Grenoble CEA Nuclear Research Center.

- $\alpha$  incineration: as part of the Melox project, SGN has adapted a standard incinerator to this incineration mode. The PWTF plant at Tokai Mura (Japan) was first equipped with an incinerator of this type by Hitachi-Zosen, SGN's licensee in Japan.

At the same time, a two-step alpha incineration process is under construction at the Valduc Nuclear Research Center with small capacity units to be operated in gloveboxes.

- For the incineration of solvents and gas, a combustion tunnel has been designed for COGEMA on the basis of an existing facility.

#### **4. 1 Standard incinerators**

These compact units feature the following components inside a sealed refractory-lined steel casing:

- a loading chute, its inlet controlled by a sealed lock with double sliding doors,
- a combustion chamber with grates designed to incinerate plastics,
- two burners, including one for post-combustion,
- a Y-shaped post-combustion chamber lined with silicon carbide aggregate, and
- an exit duct for flue gases.

This incineration unit, designed for small capacities (10 to 150 kg/h) incorporates a fume treatment system generally including:

- gas dilution by cold air,
- dry filtration (bag filters for example),
- HEPA filtration,
- if necessary, fume acidity treatment,
- gas scrubber, and
- discharge stack.

#### **4. 2 MELOX incinerator**

##### **4. 2. 1 Objectives and basic options**

The objective is to obtain maximum reduction of both the volume and the alpha activity of wastes intended for geological disposal.

The basic options retained for the management of combustible wastes resulting from the fabrication of MOX fuels are as follows:

- incineration in the MELOX plant located on the MARCOULE site, and
- treatment of plutonium-rich ashes in the La Hague UP2-800 reprocessing plant enabling:
  - the recovery of plutonium in the ashes by electrolytic dissolution and its recycling in the plant purification units, and
  - the vitrification of treated ashes.

##### **4. 2. 2 Incineration**

The combustible alpha wastes will be incinerated in an advanced version of a field-proven grate furnace, operating with excess air and provided with a postcombustion chamber filled with silicon carbide aggregates.

These wastes have specific characteristics:

- a plutonium content requiring highly reliable containment and the implementation of a criticality control system,
- a very high proportion of PVC which requires particular precautions against hydrochloric corrosion, and
- a high proportion of chloroprene with high zinc concentration which leads to an important production of volatile chlorides and requires the implementation of a specific gas treatment.

Compared with existing facilities, the MELOX incinerator whose capacity is 20 kg/h, which will start operation at the end of 1993, will be provided with miscellaneous innovations, taking into account these specific requirements.

The containment of radioactive materials in the incinerator is ensured by means of two successive static barriers strengthened by a double dynamic barrier:

- a first welded casing constitutes the first static barrier, and
- a second welded casing constitutes the second static barrier.

The space between the two casings is ventilated in excess pressure with respect to the combustion and postcombustion chambers, and under negative pressure with respect to the local environment.

The non-contamination of the air extracted from this double jacket is permanently controlled.

Particular precautions are taken in the design of casing penetrations and of the intermediate space.

The criticality control method retained consists of maintaining the plutonium mass below a limit fixed by safety authorities in each part of the facility:

- waste preparation,
- incineration, and
- ashes conditioning.

The incinerator control system is comprised of:

- permanent neutron counting around the ash removal hopper,
- neutron counting around the postcombustion chamber which takes advantage of the moderating power of silicon carbide aggregates, and
- periodical inspections and cleaning through double doors.

The refractory thickness and the double jacket ventilation are designed so that the whole first casing be maintained at a temperature higher than the maximum dew point of hydrochloric vapors.

For this the double jacket is divided into channels, the distribution of air and the temperature of which have been determined using a computerized design model.

The filtering of fumes containing zinc chloride requires prefiltration at a relatively low temperature on a cleanable filter. High efficiency bag filters have been selected.

The development tests carried out by CEA enabled the best filtering medium to be selected and the filtration and declogging parameters to be optimized.

These choices were corroborated by an endurance test performed on a representative pilot unit.

#### 4.3 VALDUC incinerator

This incinerator based on the IRIS process (pyrolysis and calcination in two rotary kilns) and built by USSI, will be operated in 1995 and will have a 7 kg/h capacity. The two-step IRIS incineration process has been tested on an inactive pilot unit of a 4 kg/h capacity which cumulates more than 3500 hours of operation.

The wastes previously crushed are pyrolysed at 550°C in an electrically heated rotary furnace. The solid residue is calcined at 900°C in a second electrically heated rotary furnace.

The corrosion tests carried out have enabled the selection of the materials best adapted to each process step and the tests performed on the pilot unit under representative conditions have validated these choices.

The tightness systems are derived from those used in the fission product vitrification furnaces of reprocessing facilities.

The furnaces are installed and maintained in glove boxes like the electrically heated rotary furnaces used for calcining plutonium oxalate in reprocessing facilities.

Pyrolysis gases are burnt at 1100°C in a fixed electrically heated postcombustion chamber lined with very high quality refractories.

The heating system design enables easy maintenance in a glove box.

After postcombustion, the fumes loaded with zinc chloride are purified on high efficiency bag filters like in the MELOX incineration system.

#### 4.4 Combustion tunnel (for liquids and gases)

For the treatment system, the method employed is excess-air combustion of the gases or liquids to be eliminated.

The system described below features superior incineration efficiency. Results achieved are ten times greater than the values specified.

An industrial incineration facility based on that technology has been operating for three years with highly satisfactory results.

##### Combustion chamber

The combustion chamber, excluding the postcombustion chamber, is sized to accommodate a heat density up to 300 thermies per hour and per cubic meter at the maximum rate of heat release.

The combustion chamber is cylindrical and horizontal in shape.

##### Burner

Aside from the combustion chamber size and the shape of the flame exit, the prime factor determining complete combustion is the quality of the fuel/oxidizing substances mix. Special measures are therefore taken to ensure high velocities at convergent jets of air from the burner firing block, whatever the flow of gas to be incinerated.

The basic arrangement for the burner and its firing block is therefore as follows:

- a central input of natural gas or liquid fuel coupled with primary air flow ensuring combustion of this gas or liquid with very small excess oxygen,
- an input of secondary air in the form of peripheral jets converging on the main burner flame. Its flow is regulated according to requirements,
- a scavenging air input at a constant flow and hence constant injection velocity to ensure a high turbulence and cooling of the firing block in all regimes, and
- inputs of gas between the secondary air and the flame of the natural gas burner via three-way valves.

##### Combustion aid

The central burner is fed constantly and the flame, monitored to ensure safety. This is carried out to control combustion of the feed gases and, if necessary, to maintain temperature in the combustion chamber.

## Temperature

The final criterion for ensuring effective combustion is the temperature inside the chamber.

The target temperature in the postcombustion chamber (regulated temperature) is 850 to 1,100°C. This temperature is slightly lower than the temperature in the combustion chamber, which is displayed in the control room.

Temperature in the postcombustion chamber is regulated by adjusting the natural gas or liquid fuel and secondary air flows using a split-range control concept.

If the temperature is greater than the set point, the natural gas or liquid fuel and primary air flows are decreased. If the temperature difference continues, the secondary air flows are increased.

## Postcombustion chamber

A second chamber filled with porous silicon carbide pebbles is placed at the combustion chamber outlet. This postcombustion chamber further enhances combustion quality by increasing the possibility of gases and unburnt solids mixing with the oxygen of the flue gases, which contain a large fraction of excess air.

The pressure drop in the postcombustion chamber is measured before and after the silicon carbide and displayed locally. This gives information about the clogging of the silicon carbide bed by dust particles.

## Conclusions

In addition to the expertise we have acquired in the implementation of beta-gamma waste incinerators with capacities up to approx. 1 T/h, two industrial incineration processes of chlorinated solid wastes highly contaminated in alpha emitters have been developed in France:

- the two-step incineration process enables safe installations in glove boxes for capacities ranging from some kg/h to 20 kg/h, and
- the direct incineration process in a high-tech fixed furnace derivated from SGN standard incinerators enables safe installations for capacities ranging from 10 kg/h to 50 kg/h.

Moreover, SGN had the original idea of installing two combustion tunnels with associated equipment in order to burn poisonous explosive gases (arsine, phosphine, silicon tetrahydride, hydrogen, etc.). They have been operating since three years with outstanding elimination efficiency (above 99.95%). This idea is now being adapted to pyrolysis gases arising from spent fuel reprocessing plants.

# **EXPERIMENTAL STUDY ON PYROLYSIS INCINERATION PROCESS FOR RADIOACTIVE WASTES**

MA Mingxie QIU Mingcai WANG Peiyi  
ZHOU Lianquan LIU Xiaoqin

China Institute for Radiation Protection

## **ABSTRACT**

In order to treat combustible radioactive wastes containing plastics and rubber in a considerable amount, a pyrolysis incineration process has been developed. Laboratory study and pilot test for the technology were performed. The results obtained in pilot test show that the waste containing a larger amount of plastics and rubber can be burnt perfectly in given technologic conditions, with a high volume-reduction factor obtained, and the process is easy to control.

### **1. Introduction**

Experimental studies have been carried out to develop an incineration process for treating uranium-contaminated wastes. The wastes to be treated contain, besides paper and cloth, a great portion of synthetic materials such as plastics, rubber, and ion-exchange resin. The process developed is pyrolysis incineration. On the basis of laboratory study, a pilot plant has been constructed, and cold test, performed. The purpose of cold test is to demonstrate the results of the laboratory study and obtain the information and data applicable for scaling-up. A continuous operation of 120 h was performed with cumulative operation time nearly 1000 h.

### **2. Principle**

The combustion behavior of cellulose materials such as cotton, paper, etc. is quite different from that of synthetic materials such as plastics and rubber. The former can be readily burnt and the latter fumes heavily when burning. The difficulty of burning the waste containing synthetic materials lies mainly in the flue gas cleaning because of the large amount of soot and tar produced in incomplete combustion. In our opinion, the rational way is to try to assure perfect combustion in order to inhibit the production of soot and tar, rather than to remove them by post-combustion or dust collecting process. This is the reason why we chose pyrolysis incineration for these wastes. In the process, the pyrolysis of waste and the combustion of volatile pyrolytic products proceeded separately. The feature of the process was to convert the direct combustion of solid materials into that of gas and tar mist which is liable to proceed perfectly so as to inhibit the formation of incomplete combustion products and simplify the cleaning of flue gas.

### **3. Process Description**

The pilot test was limited to the processes of pyrolysis, combustion, and flue gas cleaning, excluding waste pretreatment and ash handling. Shredding and packaging of waste were performed manually. The waste bags had the size of 60~80 mm. The simulated wastes were

composed of various uncontaminated materials. The typical waste composition was as follows:

cotton and paper	40%
PVC	25%
PE	15%
rubber	5%
ion-exchange resin	5%
poly-perchloroethylene	10%

A simplified flow diagram is shown in Fig. 1.

The throughput capacity of the pilot plant was nearly 4 kg/h.

### Pyrolysis

The pyrolyzer was a shaft cylindric furnace made of carbon steel without refractory lining. It was 273 mm in outer diameter and 1,100 mm in height. Waste bags were fed into the furnace batchwise and thermally decomposed into volatile products and pyrolytic char in an oxygen-deficient atmosphere. The heat required for pyrolysis was supplied by the heat evolved from the combustion of pyrolytic char left underneath with primary air. The volatile pyrolytic products, including combustible gases and tar mist, were carried away with the upward gas stream and left the furnace. The ash was swept down through the grate with a rotating rod. Its residual carbon content was below 5% (wt). The volume reduction ratio was 35 : 1 approximately.

There were jackets at the upper and lower parts of the furnace. Cooling air was blown into the lower one to cool the cylinder wall of the lower part of the furnace and thus reduce its corrosion. The hot air leaving the lower jacket entered the upper one for maintaining a higher temperature of the wall to prevent or reduce the condensation of tar mist.

The primary air amounted only to about one fourth of the stoichiometric quantity of air required for complete combustion. The low rate of air stream diminished the entrainment of ash, resulting in a low fly ash content in flue gas. Pyrolysis rate was not quite steady; during a feeding period, it was faster at the beginning and then became slower gradually. Wastes of different composition showed nearly the same behavior in pyrolysis.

### Combustion

The volatile pyrolytic products from the pyrolyzer, after mixed with sufficient secondary air, entered the combustion chamber via a nozzle. The horizontal combustion chamber was made of a high alumina ceramic tube (150 mm I. D., 750 mm L) with electroheating coil and thermal insulation outside. Under condition that the temperature in combustion chamber was above 1170K and the oxygen concentration, above 5%, perfect combustion could be obtained. The flame was clear and no smoke was perceived. The combustion temperature was commonly within the range of 1220~1420K. Normally, the combustion heat was sufficient to maintain the temperature within the range. External electroheating was supplied only when the temperature fell down below 1170K. As mentioned above, the pyrolysis rate varied in each feeding period, the oxygen concentration in flue gas varied accordingly in the range of 5~15%. It could be controlled by adjusting the secondary air flow rate.

### Cooling

The hot flue gas from the combustion chamber was cooled to 570K with an air-to-gas jacketing heat exchanger before entering the cleaning system.

## Cleaning

The flue-gas cleaning system consisted of a filter for particulate removal and an absorption scrubber for acidic gases removal. If the filtration efficiency was high enough, only particulates of very small size could escape from filtration. It was anticipated that the small particulates would not probably be removed effectively in scrubbing, thus the exhausted scrubbing solution should not be managed as radioactive liquid waste. The arrangement of the cleaning system was based upon that assumption.

Bag filter was selected for particulate removal. Filtration had to be carried out at a temperature higher than the dew point of flue gas to prevent acid condensation. The dew point of flue gas was estimated to be about 405~420K. In operation, the filtration temperature was kept above 435K. The filtration medium was NOMEX needle felt with mass thickness 350 g/m<sup>2</sup>. The face velocity was about 0.9 m/min.

The content of particulates and their size distribution were determined (see Table 1).

It can be seen from the table that for the waste containing high content of PVC and rubber the particulate contents before and after filtration are much greater than those for the waste containing only cotton and paper. In the case of the waste with high content of plastics and rubber, the results of chemical analysis showed that the particulate samples collected before and after filtration contain a great amount of Pb and Zn. These constituents are considered to be originated probably from the additives in plastics and rubber. Lead and zinc would sublime in the form of chlorides in pyrolyzer and are then converted into oxides or other compounds in combustion chamber. In cooling process of flue gas, they are condensed gradually and collected in filtration. The high content of particulates in flue gas behind filtration suggests that condensation of some sublimate could happen after filtration. Generally, the decontamination efficiency is different from the filtration efficiency. The actual decontamination efficiency will be determined using contaminated waste in further experiment.

After filtration, flue gas entered an absorption tower which was packed with ceramic corrugated plate packings (2,000 mm high). The tower was 150 mm in diameter and 4,000 mm in height. The gas temperature at the entry was about 343K. The acid gases were removed from the gas stream by countercurrent contact with alkaline solution (5% sodium carbonate solution). The concentrations of acid gases and the absorption efficiencies are given in Table 2.

Since the results of HCl removal are not satisfactory, further improvement is required.

Whether the exhausted scrubbing solution can be managed as non-radioactive liquid waste remains to be demonstrated in hot test.

A negative static pressure is maintained in entire system with an induced draft fan.

## Corrosion problems

All devices and tubes are made of carbon steel. Because of the high acid content in flue gas (see Table 2), corrosion is a serious problem. No quantitative results can be given. By visual observation, the corrosion of pyrolyzer is not serious. In the cooling and cleaning systems, the most serious corrosion occurred in the duct sections adjacent to combustion chamber and between the baghouse and absorption tower. The corrosion of devices and ducts in the temperature range of 440~680K was found to be comparatively much slighter. These results agree basically with

those given in literatures.

## Summary

In general, the results of cold test conform to those of laboratory study. The results obtained show that the process can incinerate wastes with high content of synthetic materials. Other features include low energy consumption and full combustion.

Table 1 Content and Size Distribution of Particulates

Composition of waste	Particulate content mg/m <sup>3</sup>		Percentage of different sizes(μm) (in number) *					
	before filtration	after filtration	<1	1~2	2~5	5~7	7~10	>10
Cloth, paper 100%	32	2.1	73.6	14.3	9.76	0.34	0.67	1.35
Cloth, paper 60% PVC, rubber 40%	620	39	55.0	23.8	15.3	2.0	2.5	1.33

\* before filtration

Table 2 Results of Absorption

Acidic gas	Velocity (m/sec)	Liquid rate (m <sup>3</sup> /h · m <sup>2</sup> )	Concentration of gas mg/m <sup>3</sup>	Absorption efficiency (%)
			original	after absorption
HCl	0.5	28	$1.08 \times 10^4$	$1.61 \times 10^2$
SO <sub>2</sub>	0.5	28	$4.84 \times 10^2$	1.00

1. Air heater
2. Air lock
3. Pyrolyzer
4. Cooling air
5. Feed
6. Combustion chamber
7. Radiation heat exchanger
8. View glass
9. Cooling air
10. Jacketing heat exchanger
11. Compressed air
12. Baghouse
13. Impulsion dust collector
14. Absorption tower
15. Tank
16. Pump
17. Blower
18. Discharge
19. Secondary air
20. Primary air
21. Ash pit
22. Ash

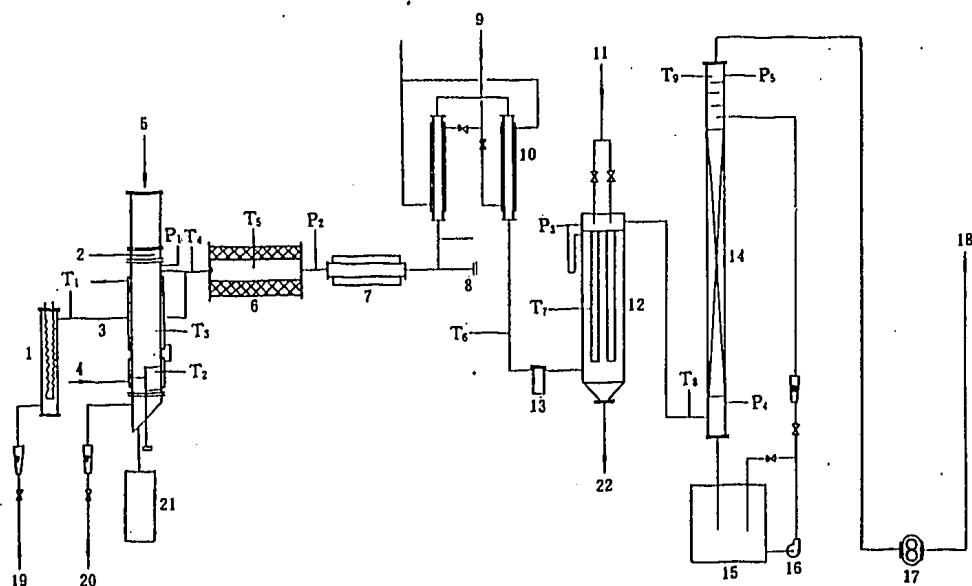


Fig. 1 The flow sheet of pyrolysis incineration

P—manometer; T—thermometer.

# MELT REFINING OF URANIUM CONTAMINATED COPPER, NICKEL, AND MILD STEEL

REN Xinwen LIU Wencang ZHANG Yuan

China Institute for Radiation Protection

## ABSTRACT

This paper presents the experiment results on melt refining of uranium contaminated metallic discards such as copper, nickel, and mild steel. Based on recommended processes, uranium contents in ingots shall decrease below 1 ppm; metal recovery is higher than 96%; and slag production is below 5% in weight of the metal to be refined. The uranium in the slag is homogeneously distributed. The slag seems to be hard ceramics, insoluble in water, and can be directly disposed of after proper packaging.

### 1. Introduction

During the processes of uranium mining and milling, uranium isotopes separation, or nuclear fuel fabrication, a large amount of spent metal parts contaminated by uranium compounds will be produced, particularly in case of decommissioning of nuclear facilities. Generally, they will be decontaminated at the original sites, then disposed of as radioactive wastes or reused as clean metals depending on their contamination levels. In order to reduce the waste volume and recover the reusable metals, melt refining can be regarded as a sound solution.

The experiment aims at selecting the suitable flux composition and process parameters, which are important factors in transferring uranium from melt into slag in addition to free energy of oxide formation.

### 2. Experiment Methodology

In order to obtain the results for refining the contaminated metal, following experiments were performed with nickel.

#### Method 1

The uncontaminated nickel was weighed and placed into crucible. Then, a designated amount of uranium compound and certain percentage of flux were added into it. After preheating, it was transferred into a resistant furnace for melting. After a certain period of time, the melt was casted. The ingot was drilled to take samples and the drilled chips of surface layer were removed. The uranium contents were measured with spectrophotometric analysis.

#### Method 2

The real contaminated nickel was weighed and placed into crucible. The flux was added with the same percentage as in Method 1. After preheating, it was transferred into resistant furnace for melting with the same temperature and duration as Method 1. Then the melt were casted. The ingot was drilled to take samples and the uranium contents were measured.

Figure 1 illustrates the results. There is the same tendency for the two curves. However,

Method 1 seems to be more sensitive to the flux amount, it is adopted for this experiment.

### 3. Results and Discussion

The thermodynamic parameters of copper, nickel, and steel are showed in Table 1 for reference.

#### 3.1 Flux selection

Each sample to be melted is 100 g by weight, in which contamination levels are 238.2 ppm, 342.3 ppm, and 238.2 ppm uranium for Cu, Ni, and mild steel respectively. Flux is added into the crucible in 10% (wt). After preheating, the mixtures are melted in a resistant furnace at different temperatures (Cu—1300°C, Ni—1550°C, steel—1600°C) for 0.5 h.

The results are listed in Tables 2, 3, 4, and Figure 2. They indicate that the decontamination efficiency will be affected by flux composition obviously. Figure 2 shows that the decontamination efficiency has a close linkage with the basicity of flux. Under the experimental conditions, better decontamination efficiency can be obtained within the basicity range from 1 to 1.3. The slag produced from blast furnace in the Taiyuan Steel Plant has a composition of 38.1 SiO<sub>2</sub>, 41.4 CaO, 3.8 MgO, 2.6 Fe<sub>2</sub>O<sub>3</sub>, and 14.1 Al<sub>2</sub>O<sub>3</sub>. Because its basicity is about 1.1, the highest decontamination factors for copper, nickel, and mild steel can be obtained in the experiment. This flux is rich in resources and low in cost. So it has been tested in more detail.

#### 3.2 Selection of melting temperature

The blast-furnace slag was selected as flux with an amount of 10% (wt), and it was melted for 0.5 hr under different temperatures. The results are showed in Figure 3. It can be found that when copper was melted above 1300°C, the temperature had little effect on decontamination efficiency. If the temperature was lower than 1300°C, ingot casting would have some difficulties. For nickel, the temperature over 1500°C also had little effect on decontamination efficiency. When the temperature was below 1550°C, ingot casting would also have some difficulties. Because of the limit of experimental furnace, 1550°C was chosen as the melting temperature for nickel.

Experimental phenomena indicate that melting temperature for decontamination should be 200~300°C above the melting point of relevant metals.

#### 3.3 Selection of melting time

Under the conditions of selected melting temperatures (Cu—1300°C and Ni—1550°C), flux type (blast-furnace slag), and the amount of flux (10% by weight), melting time was changed from 0.1 to 1 h. The results are showed in Figure 4. It indicates that extending the melting time seems disadvantageous in removing uranium from copper and no effect is found on decontamination of nickel within 0.17~1 h.

Since uranium contaminants can be extracted into slag rapidly, the metal should be casted as soon as it was fully melted. The melting time should usually not exceed 0.5 h.

#### 3.4 Effect of flux amount on decontamination efficiency

Under the conditions of selected melting temperatures (Cu—1400°C and Ni—1550°C), flux type (blast furnace slag), and melting time (0.5 h), melt refining was carried out within the

scope of 0~10% (wt) of flux amount. The results illustrated in Figure 5 show that the minimum amount of flux added should be 5% (wt), otherwise the residue contents of uranium in the ingot should increase evidently. This amount is only for laboratory scale. In industrial scale the amount will be greatly reduced. It is necessary to find out the most favourable amount to be used for the furnace.

### 3. 5 Effects of contamination level on decontamination efficiency

Under the same conditions as Section 4, contamination experiments were conducted for Cu (2.4~238.2 ppm of uranium) and Ni (342.3~680.6 ppm of uranium). The results illustrated in Figure 6 indicate that the contamination level has no effect on the residue uranium contents in the ingot. Even if this level rises to 680.6 ppm, the residue uranium content in the nickel ingot is only 1.5 ppm. In fact such a high contamination level is seldom found.

There is less need of predecontamination process for metal parts contaminated by natural uranium. For the metal parts from uranium enrichment plant, much stricter predecontamination process is necessary so as to recover uranium and avoid criticality.

### 3. 6 Refining results of real samples

Samples from nuclear fuel fabrication facilities were taken to demonstrate the refining effectiveness. The parameters are as follows. Melting temperature: Cu—1400°C, Ni—1550°C, and mild steel—1600°C; melting time: 0.5 h; flux: blast furnace slag; and amount of flux: 10% (wt). The results are listed in Table 5. The refining gives sufficient decontamination efficiency, no matter how much the contamination level is.

### 3. 7 Refining parameters recommended

The refining parameters recommended are listed in Table 6. For steel, the melting temperature depends on the sort of steel. Normally it is higher than 1600°C.

### 3. 8 Metal recovery and slag productive rate

Under the conditions of melt refining parameters recommended, the metal recovery and slag productive rate have been measured. For copper, the metal recovery is  $99.9 \pm 0.3\%$ ; slag productive rate (slag weight/amount of flux added in) is  $125.7 \pm 16.8\%$ . For nickel, the metal recovery is  $96.1 \pm 2.7\%$ ; slag productive rate is  $133.6 \pm 24.8\%$ . For mild steel, the metal recovery is  $98.9 \pm 1.6\%$ ; slag productive rate is  $128.3 \pm 21.4\%$ .

Those data were obtained in laboratory scale. The slag productive rate in industrial scale will be much less than that in laboratory scale.

### 3. 9 Uranium release rate to environment during melting

During melting, uranium may escape from crucible to the environment in different compound forms. The measurement results are listed in Table 7. It indicates that the uranium release rate is very low, which will not exceed  $10^{-5}$  of the total uranium contained in the metal to be melted.

### 3. 10 Uranium distribution in slag

200 g of flux was put into the crucible. Uranium compound containing 0.4763 g uranium was then added to. It was melted in the furnace at 1550°C for 10 minutes, and then casted. After slag ingot was cooled down naturally, samples were taken from it as shown in Figure 7 to

measure the uranium contents. The results listed in Table 8 show that uranium distribution in the slag is rather homogeneous. That total uranium amount calculated with an average uranium content in the slag is 105. 1% of that added.

Samples of slag, produced during copper melting, were taken for measurement of the uranium content, the total uranium amount calculated was 25. 24 mg, 105. 9% of the total uranium contained in the melted copper.

## Conclusions

- The results obtained accord with those anticipated by thermodynamics.
- Melt refining can transfer the uranium contaminants from metal into slag effectively.
- Residue uranium contents in the ingot after melt refining mainly depend on the basicity of flux. When the basicity falls in the range of 1~1. 3, the optimum decontamination efficiency can be obtained.
- Blast-furnace slag produced in the Taiyuan Steel Plant has a basicity of 1. 1. This slag is the most effective flux for melt refining copper, nickel, and mild steel.
- Temperature of 200~300°C above metal melting point is suitable for melt refining.
- Flux addition amount should be higher than 5% of the metal by weight when the metal is melted in laboratory scale. It could be much less than 5% in industrial scale. The proper amount of flux added in depends on the melt furnace capacity.
- The shorter the melting time, the better the decontamination efficiency. As soon as the metal is melted completely, it should be casted.
- Original contamination level has no effect on melt refining results.
- Uranium distribution in slag is sufficiently homogeneous.
- Slag looks like ceramics. It is very stable in water or diluted mineral acid. It can be disposed of directly after proper packaging.
- Using parameters recommended for melt refining in laboratory scale, residue uranium content in ingot is lower than 1 ppm; metal recovery is higher than 96%, slag productive rate is around 135%, uranium release rate is lower than  $10^{-5}$  of the total uranium in the melted metals.

Table 1 Thermodynamic Parameters

Metal	Density (g/cm <sup>3</sup> )	Melting point (°C)	Specific heat (cal/g)	Fusion heat (cal/g)
copper	8. 92	1083	0. 0916	51
nickel	8. 902	1452	0. 107	73
iron	7. 869	1536	0. 1077	64. 4

Table 2 Decontamination Efficiency of Various Fluxes for Copper

Flux No.	Flux composition (wt %)	Basicity	Residue U content (ppm)	DF*
1	100BaCl <sub>2</sub>		10.7	22.3
2	20CaO 80Fe <sub>2</sub> O <sub>3</sub>		13.3	17.9
3	65SiO <sub>2</sub> 10CaO 15Fe <sub>2</sub> O <sub>3</sub> 10Al <sub>2</sub> O <sub>3</sub>	0.21	3.8	62.7
4	40NaF 60CaF <sub>2</sub>		13.0	18.3
5	30SiO <sub>2</sub> 50CaO 5CaF <sub>2</sub> 5Fe <sub>2</sub> O <sub>3</sub> 10Al <sub>2</sub> O <sub>3</sub>	1.65	3.6	66.2
6	40SiO <sub>2</sub> 30CaO 10CaF <sub>2</sub> 20Al <sub>2</sub> O <sub>3</sub>	0.76	1.8	132.3
7	100CaF <sub>2</sub>		18.3	13.0
8	Na-glass		5.7	41.8
9	50SiO <sub>2</sub> 25CaO 25Fe <sub>2</sub> O <sub>3</sub>	0.41	3.5	68.3
10	38.1SiO <sub>2</sub> 41.4CaO 3.8MgO 2.6Fe <sub>2</sub> O <sub>3</sub> 14.1Al <sub>2</sub> O <sub>3</sub>	1.1	1.0	235.8

\* DF : decontamination factor

Table 3 Decontamination Efficiency of Various Fluxes for Nickel

Flux No.	Flux composition (wt %)	Basicity	Residue U content (ppm)	DF
3	65SiO <sub>2</sub> 10CaO 15Fe <sub>2</sub> O <sub>3</sub> 10Al <sub>2</sub> O <sub>3</sub>	0.23	10.0	34.3
5	30SiO <sub>2</sub> 50CaO 5CaF <sub>2</sub> 5Fe <sub>2</sub> O <sub>3</sub> 10Al <sub>2</sub> O <sub>3</sub>	1.63	2.8	122.3
6	40SiO <sub>2</sub> 30CaO 10CaF <sub>2</sub> 20Al <sub>2</sub> O <sub>3</sub>	0.76	2.7	126.8
10	38.1SiO <sub>2</sub> 41.4CaO 3.8MgO 2.6Fe <sub>2</sub> O <sub>3</sub> 14.1Al <sub>2</sub> O <sub>3</sub>	1.1	1.0	342.3
11	75SiO <sub>2</sub> 20CaO 5Fe <sub>2</sub> O <sub>3</sub>	0.31	4.1	83.5
12	40SiO <sub>2</sub> 40CaO 20Fe <sub>2</sub> O <sub>3</sub>	1.25	1.1	311.2
13	75SiO <sub>2</sub> 25CaO	0.36	4.7	72.8
14	10SiO <sub>2</sub> 40CaO 10Fe <sub>2</sub> O <sub>3</sub> 40Al <sub>2</sub> O <sub>3</sub>	1.38	2.2	155.6
15	50SiO <sub>2</sub> 25CaO 25Al <sub>2</sub> O <sub>3</sub>	2.4	7.4	46.3

Table 4 Decontamination Efficiency of Various Fluxes for Mild Steel

Flux No.	Flux composition (wt %)	Basicity	Residue U Content (ppm)	DF
5	30SiO <sub>2</sub> 50CaO 5CaF <sub>2</sub> 5Fe <sub>2</sub> O <sub>3</sub> 10Al <sub>2</sub> O <sub>3</sub>	1.96	6.4	37.2
6	40SiO <sub>2</sub> 30CaO 10CaF <sub>2</sub> 20Al <sub>2</sub> O <sub>3</sub>	0.76	2.2	108.3
10	38.1SiO <sub>2</sub> 41.4CaO 3.8MgO 2.6Fe <sub>2</sub> O <sub>3</sub> 14.1Al <sub>2</sub> O <sub>3</sub>	1.1	1.5	216.5
11	75SiO <sub>2</sub> 20CaO 5Fe <sub>2</sub> O <sub>3</sub>	0.31	4.9	48.6
12	40SiO <sub>2</sub> 40CaO 20Fe <sub>2</sub> O <sub>3</sub>	1.25	2.5	95.3
13	75SiO <sub>2</sub> 25CaO	0.36	4.9	48.6
14	10SiO <sub>2</sub> 40CaO 10Fe <sub>2</sub> O <sub>3</sub> 40Al <sub>2</sub> O <sub>3</sub>	1.38	3.3	72.2
15	50SiO <sub>2</sub> 25CaO 25Al <sub>2</sub> O <sub>3</sub>	0.42	3.3	72.2
16	10SiO <sub>2</sub> 50CaO 10CaF <sub>2</sub> 5Fe <sub>2</sub> O <sub>3</sub> 25Al <sub>2</sub> O <sub>3</sub>	0.40	3.5	68.1
17	40SiO <sub>2</sub> 50CaO 10Fe <sub>2</sub> O <sub>3</sub>	1.42	4.3	55.4
18	25SiO <sub>2</sub> 50CaO 25Fe <sub>2</sub> O <sub>3</sub>	0.25	4.0	59.6

Table 5 Melt Refining Results of Real Samples

Metal	Copper	Nickel		Mild steel		
Furnace No.	1	1	2	1	2	3
Original U cont. (ppm)	2	298	3111	6	25	152
Residue U cont. (ppm)	BDL*	BDL	BDL	BDL	BDL	BDL
DF	>4	>6E2	>6E3	>10	>50	>3E2

\* BDL: below determination limit

Table 6 Melt Refining Parameters Recommended

Metal	Flux	Amount of flux	Melting temp.	Melting time
Copper	Blast furnace slag	5% (wt)	1400°C	0.2~0.5 hr
Nickel	Blast furnace slag	5% (wt)	1550°C	0.5 hr
Steel	Blast furnace slag	5% (wt)	>1600°C	0.5 hr

Table 7 Uranium Release Rate to the Environment at Various Temperatures

Metal	T(°C)	Furnace No.	Total U in metal(mg)	Total U in air(mg)	Esc. rate
Copper	1250	1	47.64	1.8E-3	3.8E-5
		1	238.2	2.5E-3	1.0E-5
		2	47.64	1.5E-3	3.1E-5
		3	47.64	3.5E-3	7.3E-5
	1400	1	47.64	4.3E-3	9.0E-5
		2	47.64	5.0E-3	1.0E-5
		3	95.28	1.2E-4	1.3E-6
		4	476.4	8.8E-5	1.8E-7
		5	1209	8.5E-5	7.0E-8
		1	1600	7.5E-4	4.7E-7
Nickel	1550	1	21947.9	1.3E-3	5.9E-8
		2	47.88	1.1E-3	2.3E-5
		3	47.88	4.0E-3	8.4E-6
		4	142.92	1.7E-3	1.2E-5
	1600	1			

Table 8 Uranium Distribution in Slag

Sample No.	U1	U2	U3	U4
Sample WT(g)	0.3740	0.4000	0.5553	0.3504
U. Cont. (mg/g)	2.6404	2.0344	2.5216	2.8182
Average U Cont. (mg/g)	$2.5037 \pm 0.3357$			

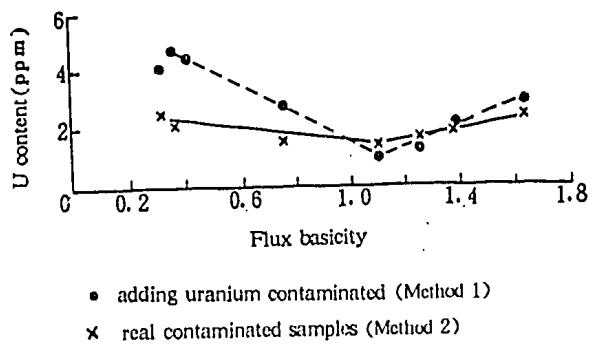


Fig. 1 Experiment method selection

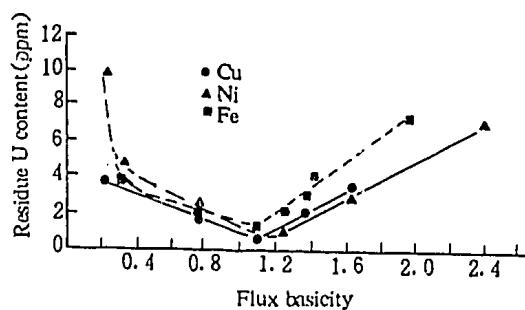


Fig. 2 Selection of flux composition

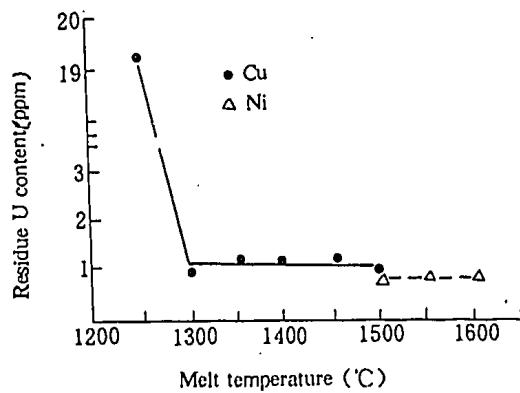


Fig. 3 Effect of temperature on residue U content

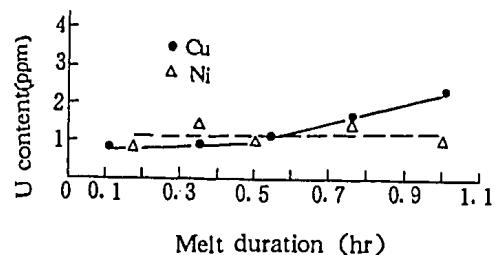


Fig. 4 Effect of melt duration on residue U content

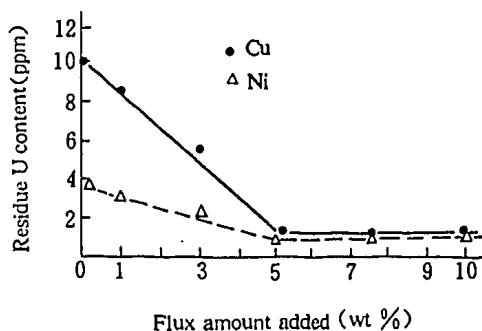


Fig. 5 Effect of flux amount on residue U content

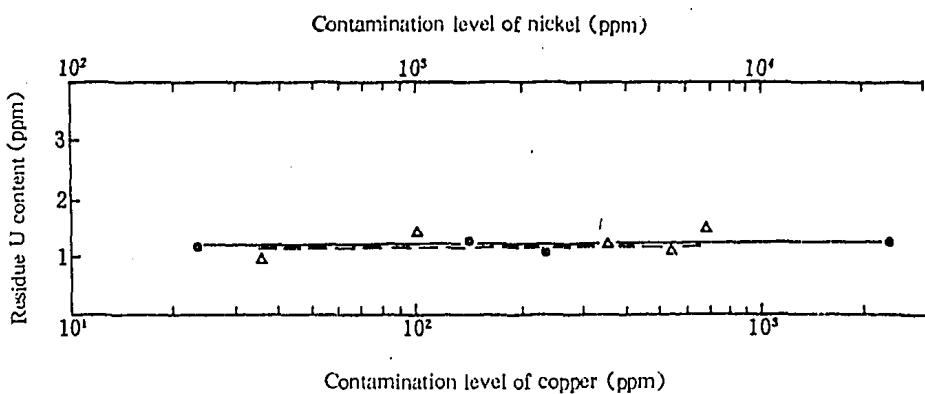


Fig. 6 Effect of contamination level on residue U content

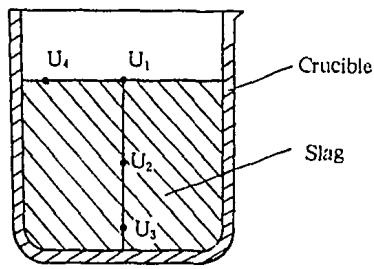


Fig. 7 Slag sampling

### References

- [1] Yemel Yanov, V. S., Yevstynkin, A. I., "The Metallurgy of Nuclear Fuel", London, Pergamon Press (1969)
- [2] Holtgren, R., "Selected Values of Thermodynamic Properties of Metals and Alloys", John Wiley & Sons Inc., New York (1963)
- [3] Heshmatpor, B., Lopeland, G. L., "The Effect of Slag Composition and Process Variable on Decontamination of Metallic Waste by Melt Refining", ORNL-TM-7501 (1982)
- [4] Abe, M., Uda, T., Iba, H., "A Melt Refining Method for Uranium-contaminated Steels and Copper", Waste Management 85, Vol. 3, pp 375~379
- [5] C. E. C., "Radiological Protection Criteria for the Recycling of Materials from the Dismantling of Nuclear Installation", DOC. No. XI-3134188 EN (1988)
- [6] LIU Wencang, REN Xianwen, ZHANG Yuan, "Micouranium Determination with TRPO-PVF2 Extrachromatographic Separation and 5-Br-PADAP Spectrophotometric Analysis", Newsletter on Radiation Protection, No. 5, 1989, p19

# MODIFICATION OF BACKFILL MATERIAL USED IN NEAR SURFACE DISPOSAL OF LOW-LEVEL RADWASTE

GU Cunli LIU Zhen FAN Zhiwen  
XIE Jianxun HUANG Yawen

China Institute for Radiation Protection

## ABSTRACT

Clay has the merits of good airtightness and strong anti-permeability when it is used as backfill material in near-surface disposal of low-level radwaste, though its ion exchange capacity is not large. In order to increase its ability to retard nuclides, 9 kinds of 5 categories of additives have been used in the test for its modification. The results show that the anti-permeability of the mixed material decreases unnoticeably, while the distribution coefficients ( $K_d$ ) for  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$  and  $^{85}\text{Sr}$  are 4.8, 4.6, and 4.7 times that using pure clay respectively, when the amount of Ca-bentonite added is 10%. The migration of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  in the clay of different densities under dynamic conditions has been studied, and the retardation factor  $R_d$ , obtained.

## 1. Introduction

As one of the engineered barriers in the near-surface disposal of low level radwaste, the backfill material can be used; (1) to minimize the flow of groundwater or surface water entering the disposal site or unit; (2) to change the chemical properties of groundwater; and (3) to retard the migration of radionuclides.

Many papers have been published about the research on the migration behavior of nuclides in backfill material. Clay and bentonite have been studied as backfill materials in many countries. "Impermeable clay" is used as backfill material in Drigg (England) and Manche (France). Groundwater movement in clay stratum of Mol Nuclear Center has been studied in Belgium, where the flowrate of groundwater is as low as  $7 \times 10^{-5} \text{ m/a}$ , the migration rate of radionuclides carried by groundwater, hence, is much slower accordingly. The mixture of bentonite and quartz sands has been studied as backfill material in Sweden, and is considered to have the properties of better adsorption and airtightness. A 30 cm thick layer of polynite/sand mixture could retard the actinide nuclides for several thousand years. These studies indicate that as a third barrier (the first is solidification waste form, the second is container), the function of backfill can not be underestimated, because a good backfill material confines the radionuclides to the vicinity of disposal site "permanently", preventing them from entering human environment.

Clay is suitable for backfill material because of its low permeability. In order to increase its retardation capability, several kinds of additives were used to modify the performance of clay. The retardation of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  was studied under various conditions through selection of additives and variation of compounding ratio of the backfill mixture;  $K_d$  value and the optimum formulations of backfill mixture were obtained. Also, the migration behavior of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  in clay columns of different consolidated densities were studied under dynamic conditions,  $R_d$  values were acquired.

## 2. Static Test

### 2. 1 Test conditions

#### (1) Selection of samples

For preventing the intrusion of groundwater effectively and sorbing nuclides strongly, the sample selected should have low permeability and high ion exchange capacity. The soil sample should be taken from the area near the disposal site to save the long-distance transportation and reduce the disposal cost. So, clay and bentonite were used in the test. Tables 1 and 2 show their chemical composition and the physical characteristics respectively.

#### (2) Pretreatment of samples

Soil samples taken from the disposal site were dried, crushed, and sieved, those with particle size smaller than 1 mm were selected as the test samples.

#### (3) Tracer solution

The solution of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  were diluted by distilled water to the predetermined concentration, then 0.1 M HCl and NaOH solution were added to adjust the pH value to about 7.

#### (4) Temperature

The test temperature was 18~20 °C, and was controlled by thermostat.

#### (5) Ratio of solid-liquid

The solid-liquid ratio of 1 : 10 (g : ml) was used in the test.

#### (6) Test Apparatus

- 25 ml polyethylene centrifugal tube with cap,
- SLJ-1 centrifuge, and
- Low background NaI  $\gamma$  spectrometer.

### 2. 2 Test method

1 g ( $1.00 \pm 0.01$ ) soil sample and 10 ml tracer solution were put into a centrifugal tube of known weight. After vibrating, dipping, and equilibrating, the sample was processed in the centrifuge with 6,000 rpm for 30 min, the upper liquid was taken out and measured with a low background  $\gamma$  spectrometer. The following equation<sup>[4]</sup> was used to calculate the distribution coefficient:

$$K_d = \frac{V}{W} \left( \frac{B}{E} - 1 \right) - \frac{X}{W}$$

Where

$K_d$  : distribution coefficient (ml/g);

$V$  : the volume of tracer solution (ml);

$W$  : the mass of solid (g);

$B$  : the radioactivity of decanted solution from blank tube (cpm/ml);

$E$  : the radioactivity of decanted solution (cpm/ml);

$X$  : the volume of residue solution after cold washing (ml).

$X / W$  of this test was 0 because distilled water was used and the cold washing of samples was not carried out.

## 2. 3 Selection of additives

The clay used in the test had good plasticity and low permeability, but low exchange capacity. For the sake of improving its retardation capability to nuclides, 9 kinds of 5 categories of additives were used in the test for modification of clay. These additives have good selective adsorption capability to Co, Cs, and Sr, and good stability under low permeable pressure. Table 3 shows the test results.

Table 3 indicates that

- (1) for the clay modified with kaolin, Na-bentonite, ZA-vermiculite, and FB-zeolite, the  $K_d$  values of the three nuclides do not increase evidently;
- (2) for the clay modified with two kinds of vermiculite, ZB and ZC, the  $K_d$  values of  $^{134}\text{Cs}$  and  $^{85}\text{Sr}$  increase evidently, but the effect on  $^{60}\text{Co}$  is not as good as that using pure clay;
- (3) for the clay modified with FA-zeolite, the  $K_d$  values of  $^{134}\text{Cs}$  and  $^{85}\text{Sr}$  increase significantly;
- (4) for the clay modified with Ca-bentonite, the  $K_d$  values of the three nuclides increase significantly; and
- (5) for the clay modified with fly ash, the  $K_d$  value of  $^{60}\text{Co}$  increases greatly; but the effects on  $^{134}\text{Cs}$  and  $^{85}\text{Sr}$  are negative.

Ca-bentonite, zeolite, and fly ash are regarded to be better additives through preliminary selection. In order to obtain the optimum formulations, the samples, to which 3 kinds of above additives of different amounts were added, were tested, the results are shown in Table 4.

Table 4 shows:

- (1) As the amount of fly ash added increases,  $K_d$  values for  $^{134}\text{Cs}$  and  $^{85}\text{Sr}$  decrease, but  $K_d$  for  $^{60}\text{Co}$  becomes higher;
- (2) When the amount of zeolite added is 5%, peaks appear for all the  $K_d$  values for the three nuclides; if the amount increases further,  $K_d$  tends to decrease; and
- (3) When the amount of Ca-bentonite added is 10%, peaks appear for  $K_d$  values for  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$ , the peak values of  $K_d$  are 4.8, 4.6 and 4.7 times that using pure clay respectively. The result shows that the mixture of Ca-bentonite and clay in ratio of 1 : 9 is the superior backfill material.

Table 5 shows the ratio of  $K_d$  of backfill mixture to  $K_d$  of pure clay of the 3 nuclides when 3 additives have optimum compositions. It can be seen from the table that fly ash has no significant effect on the three nuclides. Artificial zeolite has significant effect on  $^{85}\text{Sr}$ , but little effect on  $^{134}\text{Cs}$  and  $^{60}\text{Co}$ . Ca-bentonite has remarkable effect on all the three nuclides. Thus, Ca-bentonite is regarded to be a superior additive to modify clay, the optimum amount to be added is 10%.

## 2. 4 Determination of equilibrium time

Clay, Ca-bentonite, and their mixture were taken as the test soil samples. Tracer solutions of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  were tested separately with the 3 samples under constant temperature conditions. Table 6 and Figs. 1-1, 1-2, and 1-3 show the results.

The results show that the time for achieving adsorption equilibrium of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  is about 16 hours. It took 4 days to ensure the adsorption equilibrium to be fully reached.

It is also indicated from the results that the equilibrium time determined in distilled water is relatively short. The reason for this may be the low impurity content in distilled water.

Therefore, the determination of time for achieving equilibrium in the actual groundwater was performed. The composition of groundwater is shown in Table 7 and the time for achieving equilibrium of  $^{60}\text{Co}$  in clay, in Table 8.

It is shown from Table 8 that the time for achieving equilibrium in groundwater is about 100 hours, longer than that in distilled water. This is possibly due to the affection of coexisting ions contained in groundwater.

## 2.5 Affection of pH on $K_d$

For the sake of investigating the affection of pH on  $K_d$ , the static adsorption of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  was studied by changing the pH of solution ranging from 5 to 8. The results are shown in Table 9.

It can be seen from Table 9 that the distribution coefficients of  $^{134}\text{Cs}$  and  $^{85}\text{Sr}$  do not vary obviously in the range of pH 5~8; however,  $K_d$  value of  $^{60}\text{Co}$  increases as pH increases, this is because  $\text{Co}(\text{OH})_2$  is produced at 8 of pH which reduces the concentration of liquid phase of  $^{60}\text{Co}$ , and makes  $K_d$  value increase.

## 2.6 Affection of concentration on $K_d$

The measurement of 3 kinds of soil samples with 4 tracer solutions of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  of different concentrations was fulfilled separately, the results are shown in Table 10 and Figs. 2-1, 2-2, and 2-3.

The results show that the distribution coefficients of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  increase with the increase of solution concentrations under all the cases of 3 kinds of materials, but the increasing rate is different.

- (1) For the backfill mixture, the variation of  $K_d$ , with an increase of concentration of  $^{60}\text{Co}$  and  $^{134}\text{Cs}$ , is not large when the concentration is below  $10^{-4}$   $\mu\text{Ci}/\text{ml}$ ; and the  $K_d$  would increase evidently with an increase of concentration when the concentration is greater than  $10^{-4}$   $\mu\text{Ci}/\text{ml}$  (Figs. 2-1 and 2-2).
- (2) For the mixture,  $K_d$  increases significantly with an increase of concentration of  $^{85}\text{Sr}$ .
- (3) For pure clay and pure Ca-bentonite,  $K_d$  values of 3 nuclides increase also with an increase of concentration within the range of concentration of the test, but there is no sudden change beyond  $10^{-4}$   $\mu\text{Ci}/\text{ml}$ .

## 2.7 Conclusions

- (1) The clay modified with Ca-bentonite greatly improves the adsorption capability of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$ , when the content of Ca-bentonite is 10%; the distribution coefficients of 3 nuclides all reach peak values, thus, 9 : 1 would be the optimum ratio for the mixture of clay and Ca-bentonite in this test.
- (2) The equilibrium time increases with an increase of co-existing ions in the groundwater. The equilibrium time is 16 hrs for distilled water, and 100 hrs, for groundwater.
- (3) When the pH value ranges from 5 to 8,  $K_d$  values change unnoticeably for  $^{134}\text{Cs}$  and  $^{85}\text{Sr}$ , and obviously for  $^{60}\text{Co}$ .
- (4) For the backfill mixture, when the concentration is greater than  $10^{-4}$   $\mu\text{Ci}/\text{ml}$ ,  $K_d$  value increases obviously.
- (5) The order of 3 kinds of backfill materials affecting  $K_d$  values of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  is

mixture>pure clay>pure Ca-bentonite.

### 3. Dynamic Test

The backfill material shall be compacted in engineering practice, the compact material features higher dry volume weight and lower porosity. Its compressive strength increases and permeability decreases. As a result, its nuclide retardation capability increases. The migration behavior of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  in clay column of different compact densities has been studied.

#### 3. 1 Test apparatus

The test apparatus is shown in Fig. 3. Distilled water flows from bottle 1, which is placed at a higher level, into soil column 2; the effluent is collected in bottle 3.

Soil column, filled with compacted clay sample, is 6.18 cm in outer diameter and 4.0 cm high. It is pressed from the two ends with TFPE perforated plates. The test apparatus is placed in a thermostat, whose temperature is controlled in the range of 18~20°C.

#### 3. 2 Test procedure

The column was washed with distilled water till the effluent flowed steadily. After distributing the tracer source on the top of column uniformly, distilled water was fed in at the top of the column, and then the effluent, collected. After the test, the column was cut into thin segments when the water content in it was just the right amount. The radioactivity of soil and water samples were measured with a  $\gamma$  spectrometer.

#### 3. 3 Results and discussion

Table 11 shows the dry volume weight, porosity, and effluent of each column.

It is shown in this table that the quantity of effluent decreases with an increase of dry volume weight, the soil sample of 3# column has the largest dry volume weight of 1.57 g/cm<sup>3</sup>, and the amount of effluent is 108 ml which is the least. Accordingly, the radioactivity passing through the column also decreases with an increase of dry volume weight, namely, columns with large dry volume weight will be high in the residual radioactivity. The result of the test provides the basis for backfill engineering, namely, the backfill material should be compacted to maximum dry volume weight, so that the releasing probability of nuclides to outside can be minimized.

The percentage of residual radioactivity of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  in soil column is shown in Table 12.

It is shown in Table 12 that above 60% of radionuclides is absorbed in the first segment, whose thickness is 5.0~5.8 mm, which illustrates that clay has good adsorption capability to radionuclides. It is also shown that the residual radioactivity held in unit depth of the clay columns increases as the dry volume weight increases.

Table 14 shows the distribution of 3 kinds of nuclides in 5 columns. Figs. 4-1 to 4-5 give the vertical variations of concentrations of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{85}\text{Sr}$  along the columns respectively.

The data show that the migration behavior of  $^{60}\text{Co}$  or  $^{134}\text{Cs}$  in columns is similar, 62~83% of  $^{60}\text{Co}$  and 81~95% of  $^{134}\text{Cs}$  are absorbed on the first segment, which means that the migration rate of each kind of nuclides is slow, and the average concentration decreases rapidly with an increase of depth.  $^{85}\text{Sr}$  has a different case, the residual radioactivity of  $^{85}\text{Sr}$  on the first segment

of each column is 0 except in 1<sup>#</sup> column because of short test period, thus, the migration rate of <sup>85</sup>Sr is higher than those of <sup>134</sup>Cs and <sup>60</sup>Co in clay.

Retardation factor  $R_d$  :

In column filtration test, due to time limit, <sup>60</sup>Co, <sup>134</sup>Cs, and <sup>85</sup>Sr did not pierce through the column during dynamic test. In such case, the retardation factor  $R_d$  can be calculated by the following equation<sup>[12]</sup>:

$$R_d = 1 + K_d \frac{\rho}{n_e}$$

Where

$\rho$  : soil density (g/cm<sup>3</sup>) ; and

$n_e$  : soil porosity.

The calculated results of retardation factors are shown in Table 13.

In Table 12, the clay has higher retardation capability to radionuclides.

The retardation factor increases with an increase of dry volume weight. The retardation factors of <sup>60</sup>Co, <sup>134</sup>Cs, and <sup>85</sup>Sr of column 3<sup>#</sup> become the largest at the maximum dry volume weight, i. e. 3. 9E3, 4. 1E3 and 1. 2E2 respectively.

## Summary

- (1) When the ratio of clay and Ca-bentonite is 9 : 1, the mixture is regarded as the optimum backfill material.
- (2) As the compact density increases, the radioactivity absorbed in column increases, and the retardation factor becomes larger.
- (3) The retardation factors of 3 kinds of nuclides in clay are in the sequence of <sup>134</sup>Cs > <sup>60</sup>Co > <sup>85</sup>Sr ; for migration distance, the sequence is <sup>85</sup>Sr > <sup>60</sup>Co > <sup>134</sup>Cs.

Table 1 Chemical Composition of Backfill Material<sup>[4]</sup>

Soil No.	Soil Name	Chemical Analysis Results (%)									
		SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	FeO	TiO <sub>2</sub>	CaO	MgO	K <sub>2</sub> O	Na <sub>2</sub> O	MnO
NT	Clay	49.52	14.34	5.06	0.71	0.76	8.50	3.38	2.72	0.98	0.09
PG	Ca-bentonite	73.32	13.04	1.18	0.19	0.18	1.07	2.81	2.12	0.09	0.02
											0.04

Table 2 Physical Characteristics of Backfill Material<sup>[6]</sup>

Soil No.	NT	PG
Measuring item		
Maximum dry bulk weight (g/cm <sup>3</sup> )	1.57	1.38
Specific weight(g/cm <sup>3</sup> )	2.80	2.69
pH	8.70	5.16
Ion exchange capacity (meq/100 g soil)	17.83	72.83
Total porosity (%)	43.57	48.70
Permeability coefficient (cm/s)	$1.0 \times 10^{-8}$	$1.09 \times 10^{-5}$

Table 3 Test Results of Additive Selection ( $K_d$ , ml/g)

Nuclide		$^{60}\text{Co}$			$^{134}\text{Cs}$			$^{85}\text{Sr}$			
Additive content(%)		30	70	100	30	70	100	30	70	100	
Additives	Vermiculite	ZA	268	208	163	1720	619	518	377.7	88	29.8
		ZB	359	419	329	6813	6006	1005	329	112	75
		ZC	371	422	398	6950	6051	3624	329	166	170
	Zeolite	FA	483	393	302	6962	6712	4171	382	294	298
		FB	357	331	236	4835	4402	1089	368	220	161
	Kaolin	G	335	280	-0.4	206	338	-1.48	266	198	29
	Bentonite	PN	325	149	153	125	49	12	332	168	164
		PG	6109	4609	249	7983	2601	298	329	308	302
	Fly ash	FH	4221	3640	4045	243	92	3.5	-6	-8	-8

When clay content is 100%,  $K_{d\text{Co}} = 1760 \text{ ml/g}$ ;  $K_{d\text{Cs}} = 2670 \text{ ml/g}$ ;  $K_{d\text{Sr}} = 87 \text{ ml/g}$

Table 4 Results of  $K_d$  of Mixed Backfill Material

Amount of additive (%)	0	5	10	15	20	25	30	70	100	
Fly ash	$^{60}\text{Co}$	1760	1699	1989	3234	3385	3416	4225	3640	4045
	$^{134}\text{Cs}$	2670	1072	760	417	361	240	243	92	3.5
	$^{85}\text{Sr}$	87	74	32	28	20	21	-6	-8	-8
Ca-bentonite	$^{60}\text{Co}$	1760	7460	8520	7266	7106	6810	6109	4609	249
	$^{134}\text{Cs}$	2670	10816	12261	10176	8780	8060	7983	2601	298
	$^{85}\text{Sr}$	87	406	410	392	378	330	329	308	302
Artificial zeolite	$^{60}\text{Co}$	1760	1760	1296	1060	881	506	483	393	302
	$^{134}\text{Cs}$	2670	7355	7223	7012	6889	5643	6962	6712	4171
	$^{85}\text{Sr}$	87	661	630	573	564	415	382	294	298

Table 5 Selection of Optimum Ratio of Mixed Material

Additive	Nuclide	Optimum amount added (%)	$K_d$ Mixture / $K_d$ Pure clay
Fly ash	$^{60}\text{Co}$	30	2.4
	$^{134}\text{Cs}$	0	1
	$^{85}\text{Sr}$	0	1
Ca-bentonite	$^{60}\text{Co}$	10	4.8
	$^{134}\text{Cs}$	10	4.6
	$^{85}\text{Sr}$	10	4.7
Artificial zeolite	$^{60}\text{Co}$	5	1
	$^{134}\text{Cs}$	5	2.8
	$^{85}\text{Sr}$	5	7.6

Table 6 Determination of the Adsorption Equilibrium Time

Nuclide		$^{60}\text{Co}$									
Time (hr)		2	4	8	16	32	64	98	136	184	216
$K_d$ (ml/g)	Clay	1213	1364	1428	1826	1963	1932	1979	1982	1936	1947
	Bentonite	154	164	167	227	247	254	244	243	239	250
	Clay/Bentonite	1573	1639	4609	8708	8715	8715	8733	8728	8697	8705
Nuclide		$^{134}\text{Cs}$									
Time (hr)		2	4	8	16	32	64	98	136	184	216
$K_d$ (ml/g)	Clay	956	1282	2497	2572	2375	2453	2431	2467	2516	2488
	Bentonite	200	211	263	267	251	263	266	259	261	262
	Clay/Bentonite	1501	1630	7256	12401	12238	12249	12268	12263	12257	12256
Nuclide		$^{85}\text{Sr}$									
Time (hr)		0.5	2	4	16	27	32	40	52		
$K_d$ ml/g	Clay	91	92	90	89	89	90	92	93		
	Bentonite	14	21	26	25	24	26	24	24		
	Clay/Bentonite	273	287	470	493	504	441	475	502		

Table 7 Chemical Composition of Groundwater (mg/l)

Chemical composition	$\text{Na}^+$	$\text{K}^+$	$\text{Ca}^{2+}$	$\text{Mg}^{2+}$	$\text{Al}^{3+}$	$\text{Cl}^-$	$\text{SO}_4^{2-}$	$\text{HCO}_3^-$	$\text{CO}_3^{2-}$	$\text{NO}_3^-$	$\text{NO}_2^-$	$\text{PO}_4^{3-}$
Content	1357	29	56.7	190	0.15	1864.7	1897	56.7	3.4	37	0.06	0.1

Table 8 Total Equilibrium Time Determined in Two Systems

Nuclide		$^{60}\text{Co} \cdot K_d$ (ml/g)							
Equilibrium time (hr)		8	16	26	64	98	136	184	216
Groundwater system		98	101	131	141	173	169	169	172
Distilled water system		1428	1864	1928	1926	1979	1982	1936	1947

Table 9 Affection of pH on  $K_d$ 

Nuclide		$^{60}\text{Co}$			
pH		5	6	7	8
$K_d$ (ml/g)	Clay	660	1121	1310	2276
	Ca-bentonite	65	92	130	750
	Clay/Ca-bentonite	7580	8529	10375	11065
Nuclide		$^{134}\text{Cs}$			
pH		5	6	7	8
$K_d$ (ml/g)	Clay	1694	1765	1631	1658
	Ca-bentonite	272	248	266	282
	Clay/Ca-bentonite	12716	12579	12248	12607
Nuclide		$^{85}\text{Sr}$			
pH		5	6	7	8
$K_d$ (ml/g)	Clay	76	79	78	89
	Ca-bentonite	45	47	45	50
	Clay/Ca-bentonite	414	407	409	394

Table 10 Affection of Concentration on  $K_d$ 

Nuclide	Concentration ( $\mu\text{Ci}/\text{ml}$ )	$K_d$ ( $\mu\text{Ci}/\text{ml}$ )		
		Clay	Ca-bentonite	Clay/Bentonite
$^{60}\text{Co}$	$2.80 \times 10^{-4}$	645	62	1339
	$5.60 \times 10^{-4}$	1084	83	1573
	$2.80 \times 10^{-3}$	1874	186	4609
	$5.60 \times 10^{-3}$	2027	227	8717
$^{134}\text{Cs}$	$2.52 \times 10^{-4}$	930	108	1494
	$5.04 \times 10^{-4}$	1131	113	1566
	$2.52 \times 10^{-3}$	1595	181	7204
	$5.04 \times 10^{-3}$	2630	266	12255
$^{85}\text{Sr}$	$2.70 \times 10^{-4}$	29	3.3	30
	$5.50 \times 10^{-4}$	34	3.5	64
	$2.70 \times 10^{-3}$	75	18	243
	$5.50 \times 10^{-3}$	98	24	398

Table 11 Data about Column and Effluent

Column No.	1"	2"	3"	4"	5"
Dry Bulk Density (g/cm <sup>3</sup> )	1.47	1.52	1.57	1.54	1.51
Porosity (%)	47.5	45.7	43.9	45.0	46.11
Moving Time (day)	120	210	210	210	210
Volume of Effluent (ml)	260	115	108	115	290
Average Flowrate (E-7)cm/s	8.46	2.23	1.98	2.23	5.32
Gross Radioactivity of Effluent (E-4)μCi	66.22	9.12	1.20	2.03	29.0
Leached Percentage (%)	11.65	0.192	0.0264	0.0459	0.673
Residue Percentage in Column (%)	88.35	99.81	99.97	99.95	99.33

Table 12 Percentage of Residual Radioactivity in Soil Column

Column	Slice No.	1	2	3	4	5	6	7
1"	Slice thickness (mm)	5.0	4.5	5.0	8.0	4.0	6.5	8.0
	Residue percentage (%)	64.0	8.89	5.56	4.76	2.03	2.34	3.95
	Residue percentage (%) / mm	12.8	1.97	1.10	0.595	0.507	0.360	0.49
2"	Slice thickness (mm)	5.2	6.3	6.3	6.3	5.2	10.5	R
	Residue percentage (%)	68.1	22.0	3.02	2.50	2.60	0.637	1.72
	Residue percentage (%) / mm	13.1	3.49	0.479	0.397	0.5	0.218	
3"	Slice thickness (mm)	5.7	5.8	5.7	5.8	5.7	11.5	R
	Residue percentage (%)	80.6	13.2	1.62	2.51	2.09	1.87	1.04
	Residue percentage (%) / mm	14.1	2.31	0.284	0.433	0.367	0.253	
4"	Slice thickness (mm)	5.8	7.8	5.2	4.8	5.8	10.5	R
	Residue percentage (%)	78.9	19.8	2.37	7.97	1.82	1.66	0.619
	Residue percentage (%) / mm	13.6	2.53	0.456	1.66	0.314	0.319	
5"	Slice thickness (mm)	5.7	5.7	5.0	4.7	7.4	11.5	R
	Residue percentage (%)	77.9	7.76	2.58	2.23	3.93	4.26	1.81
	Residue percentage (%) / mm	13.7	1.36	0.516	0.474	0.531	0.527	

(R—residual soil)

Table 13 Retardation Factors for Three Kinds of Nuclides

Nuclide	<sup>60</sup> Co	<sup>134</sup> Cs	<sup>85</sup> Sr
Column 1"	3.3E3	3.5E3	1.1E2
Column 2"	3.6E3	2.8E3	1.1E2
Column 3"	3.9E3	4.1E3	1.2E2
Column 4"	3.7E3	3.9E3	1.2E2
Column 5"	3.5E3	3.7E3	1.1E2

Table 14 Results of Soil Samples Measured in Sections

Sample No.	Slice thickness (mm)	Gross radioactivity ( $\mu\text{Ci}$ )			Weight (g)	Average concentration ( $\mu\text{Ci/g}$ )			Cumulative depth (mm)	Percentage of cumulative radioactivity (%)			Average depth (mm)
		Co-60	Cs-134	Sr-85		Co-60	Cs-134	Sr-85		Co-60	Cs-134	Sr-85	
1 <sup>#</sup> -1	5.0	1.68E-1	1.77E-1	1.33E-2	26.24	6.40E-3	6.74E-3	5.07E-4	5.0	62.55	95.02	10.93	2.50
1 <sup>#</sup> -2	4.5	7.99E-3	7.90E-3	3.39E-2	23.64	3.38E-4	3.34E-4	1.43E-3	9.5	65.52	99.27	38.79	7.25
1 <sup>#</sup> -3	5.0	3.65E-3	1.15E-4	2.74E-2	26.24	1.39E-4	4.38E-6	1.04E-3	14.0	66.88	99.33	61.31	12.0
1 <sup>#</sup> -4	8.0	1.84E-3	D.L	2.52E-2	42.00	4.38E-5	D.L	6.00E-4	22.5	67.57	99.33	79.30	18.0
1 <sup>#</sup> -5	4.0	2.11E-3	D.1	9.29E-3	21.00	1.01E-5	D.L	4.42E-4	26.5	68.35	99.33	87.44	24.5
1 <sup>#</sup> -6	6.5	4.10E-3	D.L	9.10E-3	34.12	1.21E-4	6.36E-6	2.67E-4	33.0	69.88	99.33	94.92	29.7
1 <sup>#</sup> -7	8.0	1.59E-2	2.67E-4	5.98E-3	42.00	3.78E-5	1.38E-4	41.0	75.80	99.47	99.84	99.84	37.0
W		6.50E-2	9.84E-4	2.00E-4					100	99.99	99.99	99.99	
2 <sup>#</sup> -1	5.2	1.79E-1	1.48E-1	D.L	23.84	7.42E-3	6.18E-3	D.L	5.2	66.41	81.32	0	2.60
2 <sup>#</sup> -2	6.3	7.23E-2	3.37E-2	D.L	28.89	2.50E-3	1.67E-3	D.L	11.5	93.24	99.84	0	8.35
2 <sup>#</sup> -3	6.3	3.40E-3	1.59E-3	9.52E-3	28.89	1.18E-4	5.50E-5	3.29E-4	17.8	94.50	99.99	30.35	14.6
2 <sup>#</sup> -4	6.3	4.20E-3	D.L	7.84E-3	28.89	1.76E-4	D.L	2.71E-4	24.1	96.06	99.99	55.34	20.9
2 <sup>#</sup> -5	5.2	4.83E-3	D.L	7.63E-3	23.84	2.03E-4	D.L	3.20E-4	29.3	97.85	99.99	79.66	26.7
2 <sup>#</sup> -6	10.5	1.62E-3	D.L	1.44E-3	48.15	3.36E-5	D.L	2.99E-5	39.8	98.45	99.99	84.25	34.5
R		3.33E-3	2.75E-5	4.88E-3					99.68	99.99	99.99	99.80	
W		8.42E-4	8.45E-6	6.04E-5					99.99	100	99.99	99.99	
3 <sup>#</sup> -1	5.7	1.78E-1	1.69E-1	D.L	26.85	6.63E-3	6.00E-3	D.L	5.7	68.54	87.80	0	2.85
3 <sup>#</sup> -2	5.8	4.39E-2	1.09E-2	7.00E-3	27.32	2.34E-3	7.65E-4	2.56E-4	11.5	93.15	99.20	19.61	8.60
3 <sup>#</sup> -3	5.7	7.33E-3	4.36E-4	D.L	26.85	2.73E-4	1.62E-5	D.L	17.2	95.97	99.44	19.61	14.4
3 <sup>#</sup> -4	5.8	2.87E-3	1.81E-4	7.58E-3	27.32	1.05E-4	6.62E-6	2.77E-4	23.0	97.07	99.54	40.84	20.1
3 <sup>#</sup> -5	5.7	1.98E-3	D.1	8.08E-3	26.85	7.37E-5	D.L	3.01E-4	28.7	97.84	99.54	63.47	25.8
3 <sup>#</sup> -6	11.5	9.94E-4	3.75E-4	7.63E-3	54.17	1.83E-5	6.92E-6	1.41E-4	40.2	98.22	99.75	84.84	34.6
R		4.50E-3	4.62E-4	5.41E-3					99.96	99.99	99.99	99.99	
W		1.14E-4	2.64E-6	3.06E-6					99.99	100	99.99	100	

(continue)

Table 14 Results of Soil Samples Measured in Sections

Sample No.	Slice thickness (mm)	Gross radioactivity ( $\mu\text{Ci}$ )			Weight (g)	Average concentration ( $\mu\text{Ci/g}$ )			Cumulative depth (mm)	Percentage of cumulative radioactivity (%)			Average depth (mm)
		Co-60	Cs-134	Sr-85		Co-60	Cs-134	Sr-85		Co-60	Cs-134	Sr-85	
4#-1	5.8	1.94E-1	1.53E-1	D.L	26.83	7.23E-3	5.17E-3	D.L	5.8	66.76	87.82	0	2.90
4#-2	7.8	6.14E-2	2.08E-2	1.29E-2	36.04	1.70E-3	5.77E-4	3.58E-4	13.6	87.89	99.76	31.36	9.70
4#-3	5.2	4.30E-2	1.62E-4	6.05E-3	24.02	1.79E-3	6.74E-6	2.52E-4	18.8	89.37	99.85	46.07	16.2
4#-4	4.8	2.70E-3	4.31E-5	8.05E-3	22.18	1.22E-4	1.93E-6	3.63E-4	23.63	98.66	99.87	65.64	21.2
4#-5	5.8	1.43E-3	4.29E-5	6.53E-3	26.80	5.43E-5	1.60E-6	2.44E-4	29.4	99.15	99.90	81.51	26.5
4#-6	10.5	2.05E-3	3.81E-5	5.23E-3	48.51	4.23E-5	7.85E-7	1.08E-4	39.9	99.86	99.92	94.22	34.6
R		2.96E-4	1.36E-4	2.29E-3						99.96	99.99	99.79	
W		1.15E-4	2.18E-6	8.58E-5						99.99	100	100	
5#-1	5.7	1.78E-1	1.57E-1	D.L	25.82	6.89E-3	6.08E-3	D.L	5.7	77.19	93.62	0	2.85
5#-2	5.7	1.70E-2	7.17E-3	9.19E-3	25.82	6.58E-4	2.78E-4	3.65E-4	11.4	84.56	97.89	25.04	8.55
5#-3	5.0	9.75E-3	1.34E-3	D.L	22.65	4.30E-4	5.92E-5	D.L	16.4	88.78	98.70	25.04	13.9
5#-4	4.7	2.90E-3	6.27E-4	6.05E-3	21.29	1.36E-4	2.94E-5	2.84E-4	21.1	90.04	99.07	41.53	19.1
5#-5	7.4	6.50E-3	D.1	1.04E-2	33.52	1.94E-4	D.L	3.10E-4	28.5	92.86	99.07	69.86	24.8
5#-6	11.5	1.51E-2	1.37E-4	3.07E-3	52.10	2.90E-4	2.63E-6	5.89E-5	40.0	99.41	99.15	78.23	34.2
R		1.28E-3	1.42E-3	5.10E-3						99.96	99.99	92.12	
W		8.21E-5	D.L	2.89E-3						99.99	99.99	99.99	

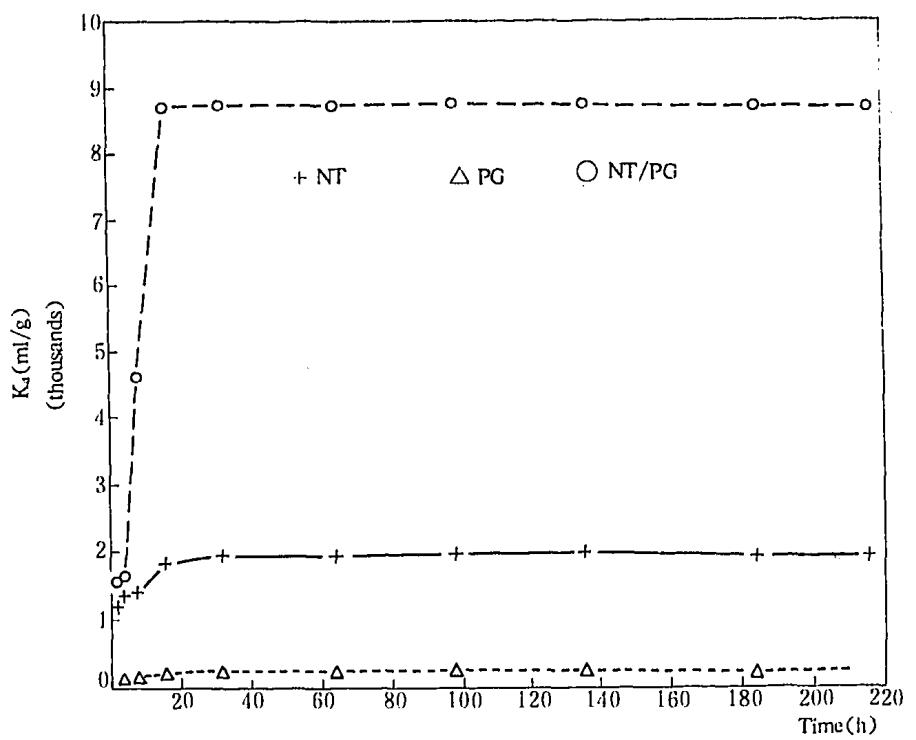


Fig. 1-1 Equilibrium curve of  $^{60}\text{Co}$  in 3 kinds of soil samples

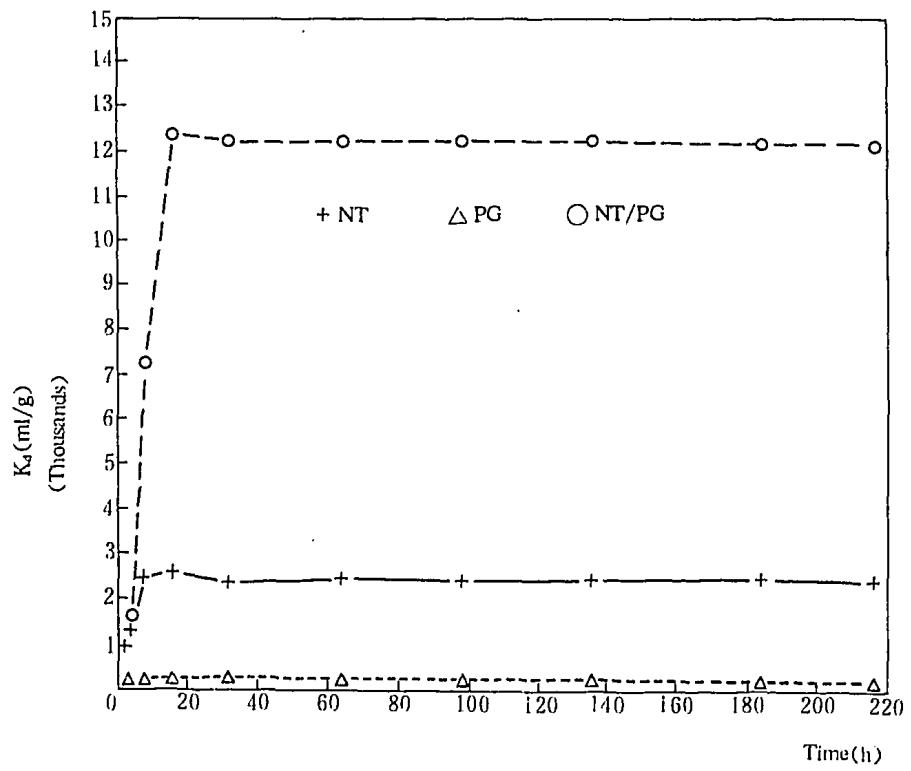


Fig. 1-2 Equilibrium curve of  $^{134}\text{Cs}$  in 3 kinds of soil samples

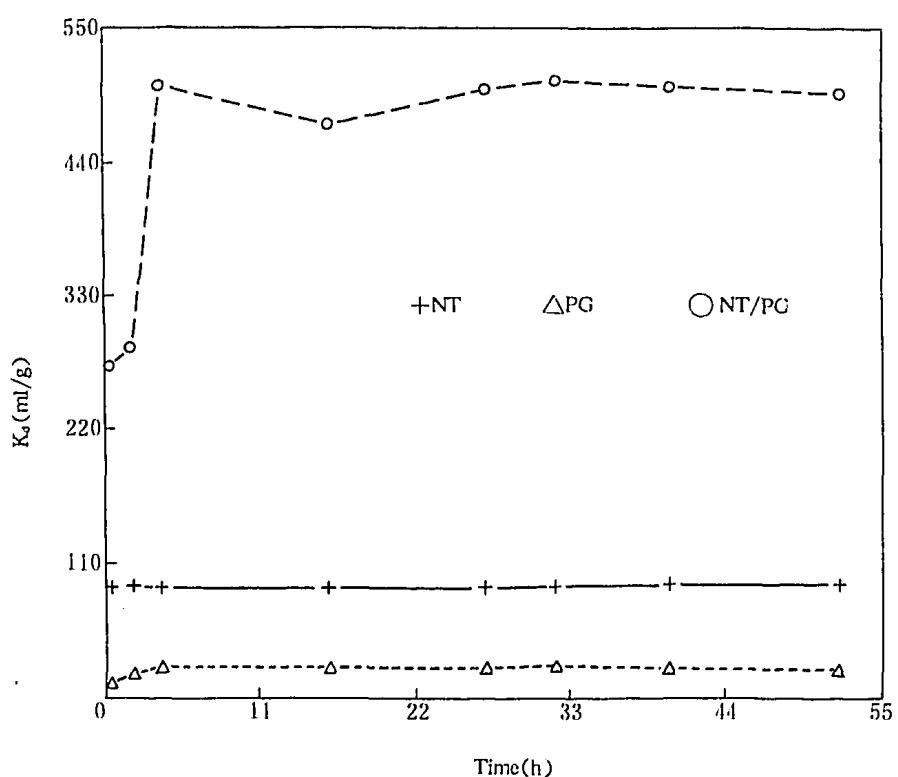


Fig. 1-3 Equilibrium curve of  $^{85}\text{Sr}$  in 3 kinds of soil samples

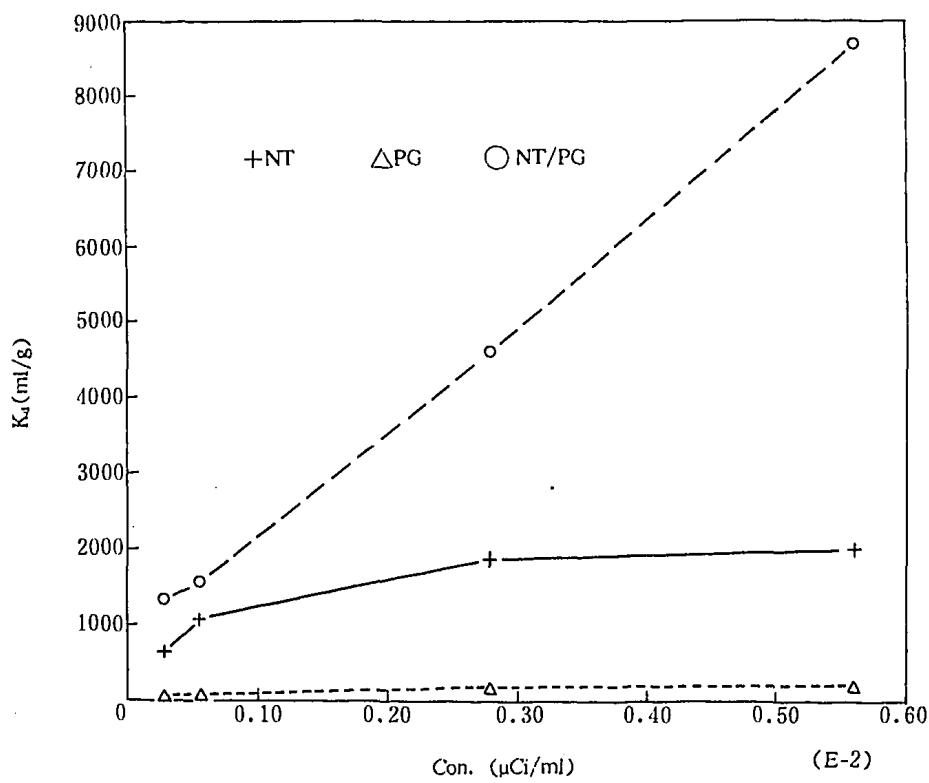


Fig. 2-1 Variation of  $K_d$  with concentration of  $^{60}\text{Co}$

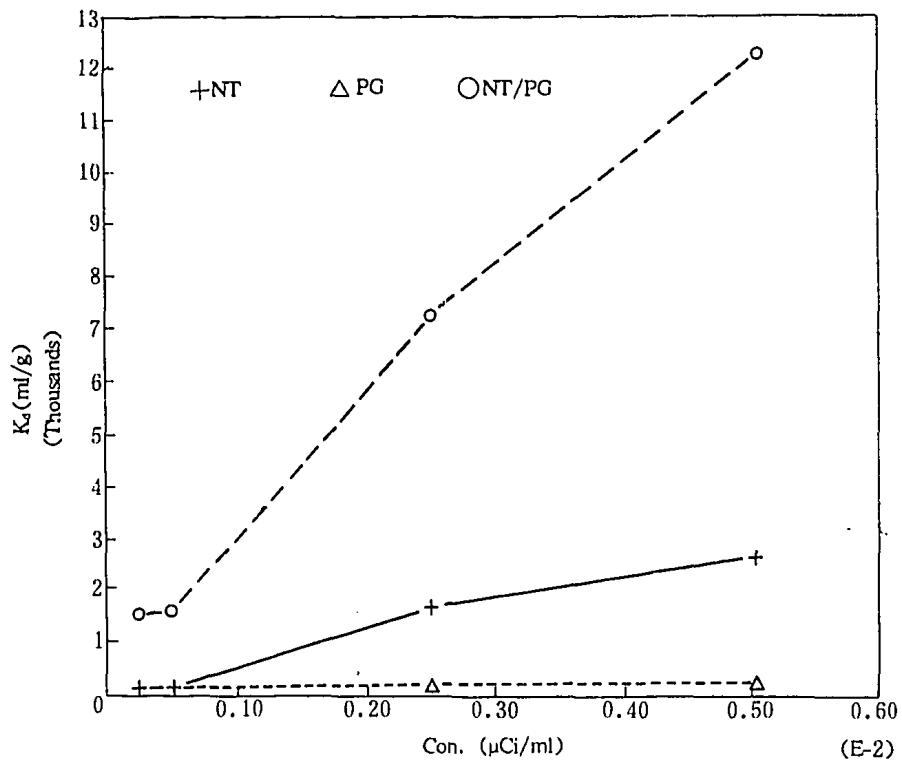


Fig. 2-2 Variation of  $K_d$  with concentration of  $^{134}\text{Cs}$

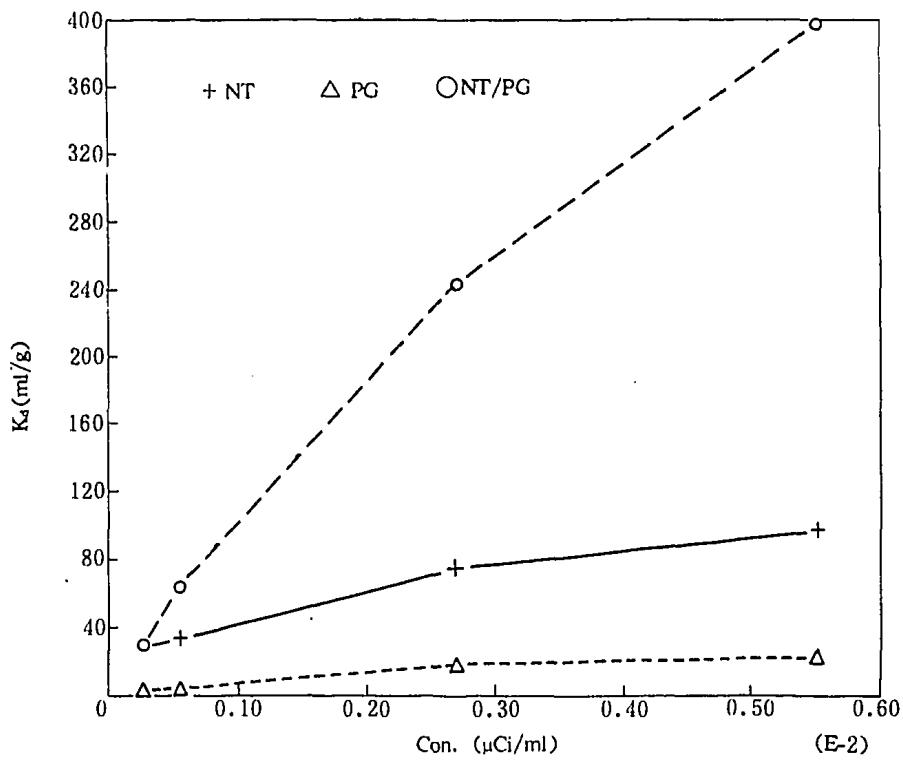
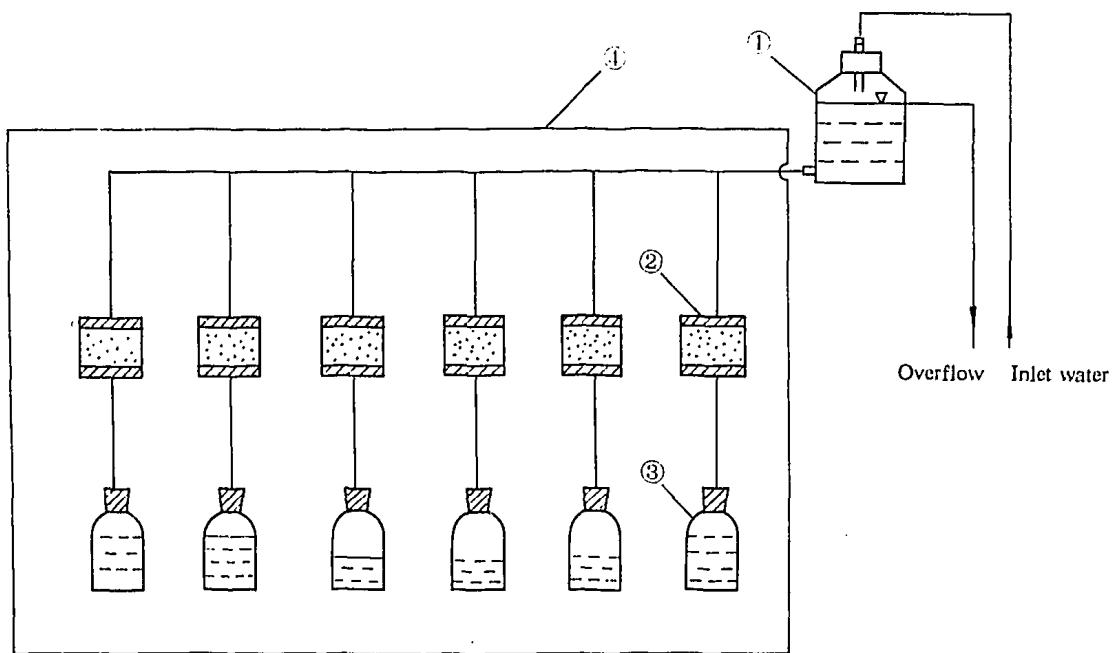


Fig. 2-3 Variation of  $K_d$  with concentration of  $^{85}\text{Sr}$



(1)High level tank of water (2)Testing clay column  
 (3)Collecting liquid bottle (4)Thermotank

Fig. 3 Schematic diagram of dynamic test system

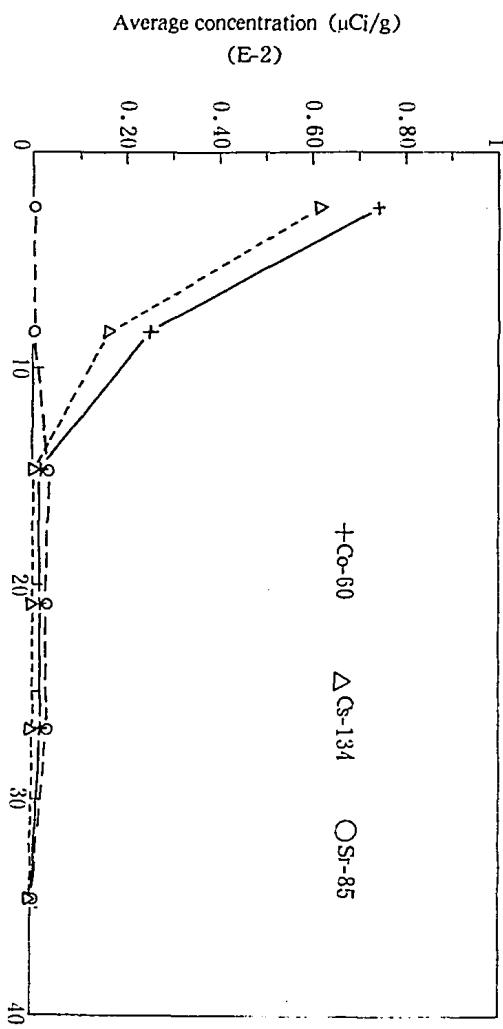


Fig.4-2 Variation of Nuclide concentration with depth in 2<sup>nd</sup> column

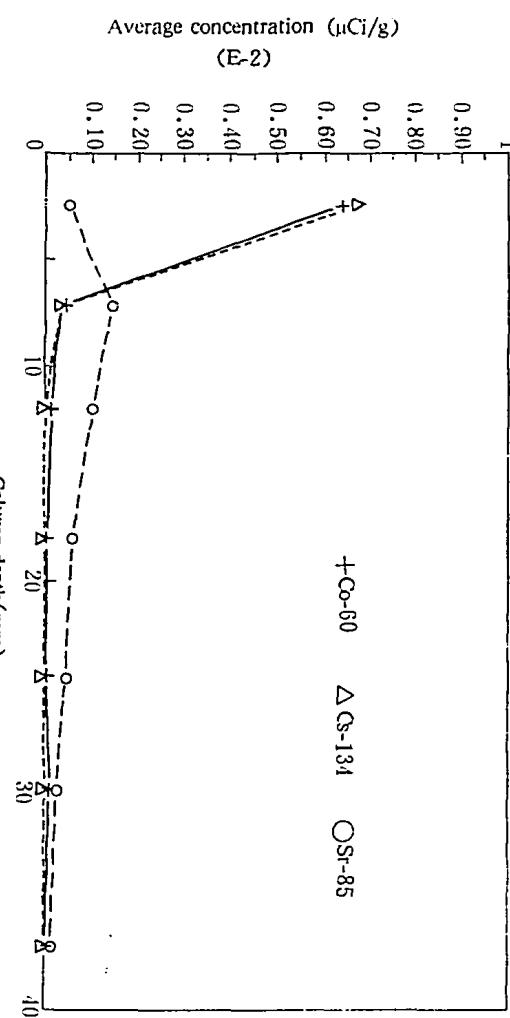


Fig.4-1 Variation of nuclide concentration with depth in 1<sup>st</sup> column

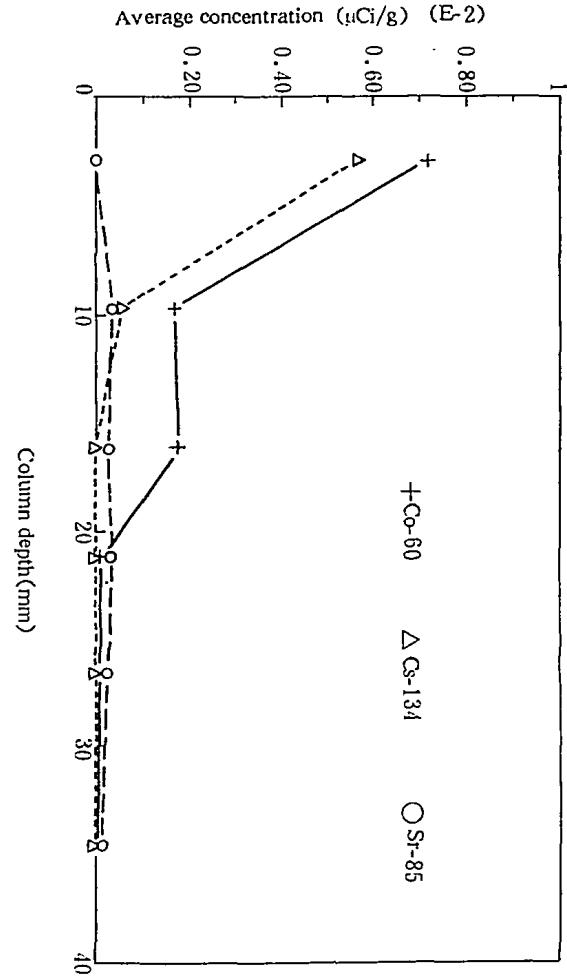


Fig. 4-4 Variation of nuclide concentration with depth in 4<sup>#</sup> column

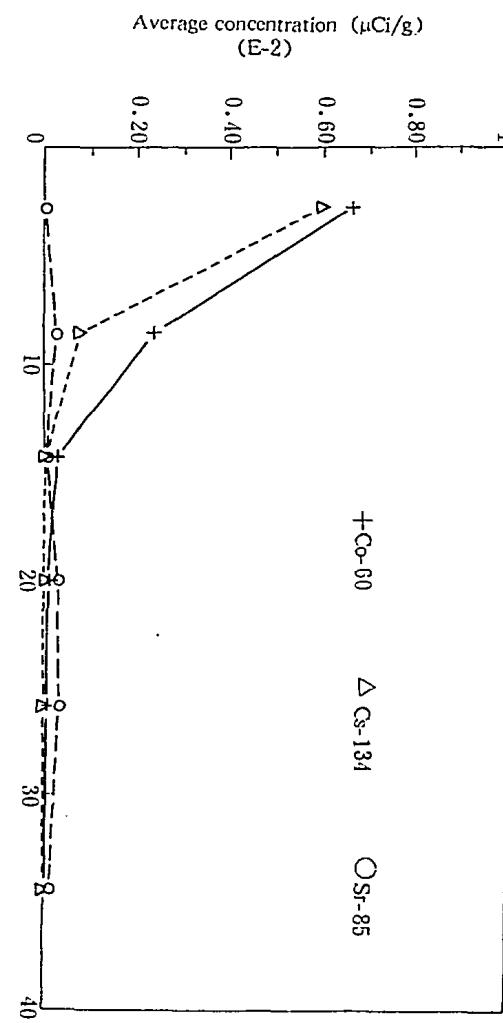


Fig. 4-3 Variation of nuclide concentration with depth in 3<sup>#</sup> column

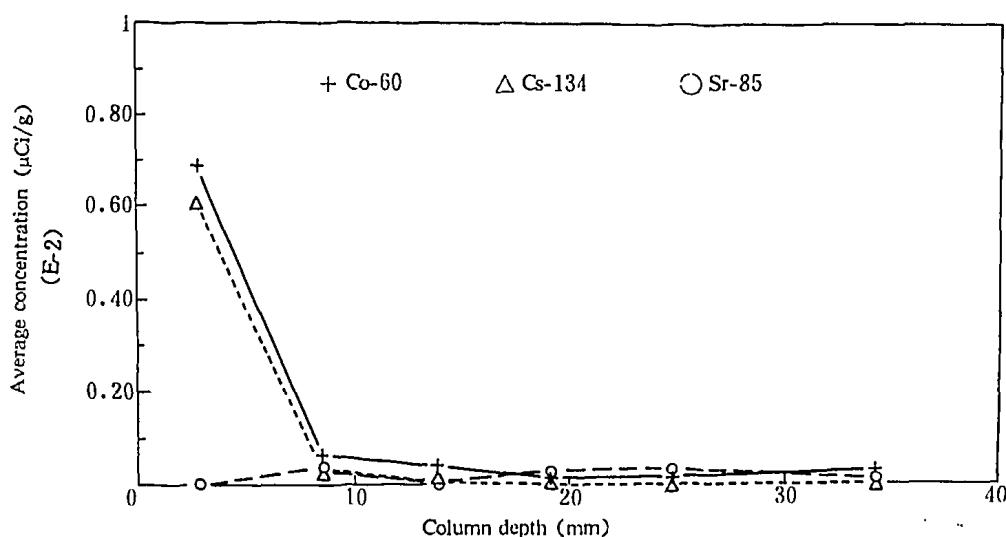


Fig.4-5 Variation of nuclide concentration with depth in 5# column

### References

- [1] Neila chapman et al. The Geological Disposal of Nuclear Waste, p. 76, 1987.
- [2] Komad. B. Krauskopf. Radioactive Waste Disposal and Geology, p. 44-53, 1988.
- [3] K. Shimooka et al. Proceedings of the 1989 Joint International Waste Management Conference Vol. P581-586.
- [4] J. K. Reyea et al. Methods for Determining Radionuclide Retardation Factors: Status Report. PNL-3349, 1980. 4.
- [5] Special Group for Radwaste Disposal of Science and Technology Committee of Ministry of Nuclear Industry Radwaste Disposal, 1983.
- [6] GU Cunli et al. Study on Backfill Material Used in Shallow-Land Disposal of LLRW (unpublished)
- [7] T. Yamamoto et al. Radioisotopes. Vol. 129, No. 8 (1980)
- [8] WANG Baozhen. Treatment of Radioactive Waste Water, Science Press, 1979.
- [9] T. Inoue et al. Japanese Nuclear Energy Society, Vol. 18, No. 8 (1976).
- [10] Handbook of Domestic and Overseas Architectural Standards (Vol. 2), Heilongjiang Sci. & Tech. Press, 1984. 4.
- [11] T. Yamamoto et al. JAERI-memo. 60-188, 1985. 8.
- [12] PAN Ziqiang. Radwaste Management, Atomic Energy Press, p81-89, 1987. 9.

# FRENCH PROCESSES FOR WASTE EMBEDDING

## THE USE OF EPOXY RESIN FOR WASTE CONTAINMENT

X. AUGUSTIN J. C. GAUTHEY  
TECHNICATOME, FRANCE

### ABSTRACT

The low- and medium- level wastes generated by nuclear facilities when operating as well as during their decommissioning (dismantling, decontamination, etc.) are embedded for the purpose of obtaining a product suitable for disposal.

Due to the varieties of waste produced, it was necessary to resort to multi-purpose techniques to solve problems relating to their embedding.

The process for waste embedding in thermosetting polymer (polyester, epoxy) developed by the French Atomic Energy Commission (CEA) and its subsidiary TECHNICATOME is easy to operate and yields excellent results having regard to volume reduction and containment of radioisotopes (particularly caesium). The industrial development of this process has led to the design of small, flexible, fixed or mobile, embedding stations. Examples illustrating the increasing use of this process during facility dismantling, are described in the presentation.

### 1. Introduction

The increasing production of radwaste coming from various operating facilities and/or resulting from dismantling obsolete facilities has resulted in the need for developing multi-purpose techniques for waste volume reduction and radioactivity containment, leading in time to proper radioactivity containment.

Accordingly the CEA has developed at Grenoble Nuclear Research Center a process for embedding LA and MA waste in thermosetting resins of the polyester type and more recently of the epoxy type.

The CEA embedding process which is available from TECHNICATOME is currently applicable to the following :

- waste from nuclear reactors and nuclear research centers: ion exchange resins (I. E. R.), powders, evaporator concentrates, sludges, and filtering cartridges, and
- waste for dismantling of contaminated facilities and/or equipment (obsolete or irreparable pieces of equipment, etc.)

The process is particularly suitable for waste containing caesium (low release rate).

Furthermore it imparts to the end product the following major advantages:

- greatly reduced volume of the embedded waste, this facilitating interim storage, transport, and disposal,
- excellent radioactivity containment: for instance in the case of embedding of spent IER, leaching rates 455 days are about  $4 \times 10^{-5}$  cm/d for caesium-137 and  $2 \times 10^{-6}$  cm/d for cobalt-60 ,
- excellent ageing resistance in particular due to adequate radiation resistance and the non-

biodegradable nature of resins in use; after integration of  $5 \times 10^{-9}$  rads, crushing strength 200 to 650 kg/cm<sup>2</sup> according to the waste, 1 mole of gas per kg of waste and matrix released by radiolysis, and

- greatly simplified operation and possible adaptation to a great number of waste types.

Extension of the process to epoxy resins has ensured the improved simplicity in processing by elimination chemical pretreatment and accepting high proportions of water — in particular for spent Ion Exchange Resins — combined with greater operating safety since no volatile solvent is used.

Cost/effectiveness calculations have shown that this process is highly competitive from the standpoints of technical flexibility, low outlay costs, and simplicity in use.

## 2. French Experience in Use of Epoxy Embedding for Dismantling Waste

Progress of the CEA nuclear facilities decommissioning program led to successive implementations of epoxy process for dismantling waste embedding purpose:

- Cs-137 and Sr-90 contaminated solid waste from ELAN 2B facility (LA HAGUE),
- main containment barrier of tritium contaminated metallic waste blocked with concrete at the research reactor EL 3 (SACLAY),
- IERs of the pool water purification system of research reactor TRITON (FONTENAY AUX ROSES), as well as IERs of the D<sub>2</sub>O and B<sub>2</sub>O circuits purification system of power reactor EL 4 (Centrale nucléaire des MONTS D'ARRES).

## 3. Fixing Contamination and Embedding Waste at ELAN 2B

### 3.1 Reasons for use of epoxys

The ELAN 2B facility (Fig. 1) is composed of a group of seven shielded cells housing the equipment of an industrial manufacturing process for sealed sources of strontium-90 and caesium-137.

Stage 3 decommissioning is presently under way involving all contaminated equipment dismantling and disposal, and final decontamination of cells, walls, and floor when necessary.

The specificity of the radionuclides present, Cs-137 and Sr-90, and the importance of the radioactivity encountered (certain cells rad ambient dose rates about 1,000 rad/h) necessitated the in situ use of a waste embedding process using thermosetting resin.

### 3.2 Painting of waste

After dismantling, a painting operation is performed to fix solid waste contamination before their transfer to the embedding cell.

The operation is carried out using an epoxy painting process which ensures good coverage of waste thanks to an electrical field applied between the paint gun and the table on which waste is placed for painting.

The electrostatic paint gun is handled by a remote-manipulator. The power supply, fluid supply, and regulation equipment, in particular the electrostatic generator, are located outside the cell.

### **3.3 Embedding of waste (Figs. 2 and 3)**

The solid waste produced by ELAN 2B is essentially composed of :

- metal fragments (piping, valves, tanks, and tools),
- filter elements, and
- various solid waste (plastics).

Painted wastes are transferred to the embedding cell, where they are arranged in a basket placed in a 220 l epoxy-resin drum which ensures suitable containment of radioelements and appropriate properties with respect to leaching. The drums are placed on a four station rotary table.

Embedding of waste is carried out in an epoxy resin charge with an inert material in order to achieve waste immobilization.

The embedding mixture is made up in a continuously running mixer. Ingredients are fed in separately. Adjustment of the dose of each the ingredients results in the desired composition of the embedding mixture. The reservoirs and the mixer are located outside the embedding cell.

The embedding mixture is delivered by a line through the wall of the cell into the drum containing the waste. When the level in the drum reaches a grid which prevents low-density material from rising to the top, a pure epoxy resin plug is poured to achieve the containment.

All the work inside the embedding cell is performed using remote manipulators.

After control, the drums are transferred via the superstructure in a shielded cask for storage to the ANDRA disposal facility.

The unit has already produced approximately one hundred drums.

One of the advantages of this process is that minimum equipment is located in the cell, which facilitates maintenance operations.

## **4. Tritium Barrier at EL3**

### **4.1 Reasons for use of epoxys**

The EL3 reactor was a research reactor moderated and cooled by heavy water. The power was 18 MWth.

Stage 2 decommissioning involved dismantling and disposal of D<sub>2</sub>O coding and auxiliary circuits. These circuits had been contaminated by tritiated heavy water, due to tritium generation from deuterium under neutron flux. So dismantling metallic waste (fragments of pipes, flanges, heat exchanger, valves, etc.) had to be suitably processed and packaged for disposal with respect to this type of contamination.

French ANDRA (Agence Nationale pour les Déchets Radioactifs) has specified standards concerning the storage of tritium contaminated waste, which refer :

- to the specific activity in tritium of the waste (tight envelope for waste with activity with respect to mass of between 0.23 Ci/t and 2 Ci/t) and
- tritium release rate.

From a practical point of view, the regulations relative to release rate appear the most restrictive. It was necessary to design a packaging process which ensures containment for the purpose

of final disposal. The epoxys were used to this end.

#### 4.2 Embedding of waste

Prefabricated cylindrical containers were used for packaging: they comprise a reinforced concrete shell, internally lined by a reinforced epoxy structure, with a 3 mm thick inner layer of pure epoxy resin, achieving the specified containment (Fig. 4).

The container has the same outer shape as standard so-called C 1 concrete containers, in order to meet disposal site requirements.

Overall dimensions are the following:

O. D. : 3,400 mm

Height: 1,300 mm

Cavity volume: approx. 1 m<sup>3</sup>.

Industrial manufacturing process includes appropriate controls of epoxy barrier.

Waste embedding operation has been performed in the PL3 reactor building.

Sequence is as follows:

After placing waste in the container, cement is poured for immobilization. A 4 mm thick layer of pure epoxy poured at the surface of the concrete ensures the continuity of the epoxy barrier with the lining of the walls, thus ensuring the containment of radio-elements, tritium in particular.

A poured, reinforced concrete lid ensures closure of the container.

#### 4.3 Pratical results

A series of twelve containers has been manufactured and fifteen metric tons of tritium contaminated waste (up to 2 Ci/t) generated by the dismantling have been embedded in the containers and disposed of. Tritium release tests were satisfactory for all packages.

### 5. Coating of Ion-Exchange Resins at Triton

#### 5.1 Justification for epoxy embedding

Triton reactor was a 5 MW open core pool type research reactor;

Stage 3 decommissioning involved dismantling of in pool equipment before draining the pool water and decontaminating the pool walls and bottom.

So the pool water purification system had to remain in operation during equipment dismantling and water draining, and spent IERs of the purification plant had to be finally disposed of in an appropriate waste form.

Thanks to their properties, developed in par 1, the epoxys were used for embedding of these resins using the SETH-200 mobile unit.

Work was performed in 1980.

#### 5.2 Description of SETH-200 and embedding sequence

SETH-200 (Fig. 5) is designed to perform IERs embedding by epoxy process in 55 gallon standard drums.

It is for a compact concept and is made of modules which can be transported by truck and easily assembled. It includes independent radiation shielding and liquid waste collecting devices.

The unit is equipped with two working stations, enabling interchange of empty and full drums while processing on the other line.

Embedding sequence is summarized as follows (Figs. 6 and 7) :

- the amount of spent ion exchange resins necessary to make a drum is transferred hydraulically into a metering pot and then into an embedding drum in its transport container ,
- after IERs draining in the drum, the reagents forming the embedding matrix are transferred from their respective dosing pots to the embedding drum,
- the constituents are then mixed with an appropriate sequence ,
- after interchanging the working places, the package is moved to a temporary storage area ,
- when the polymerization has been checked, a sealing plug is injected to close the drum , and
- after full polymerization, the package may be sent to the storage center.

The 55 gallon packages are solid, uniform blocks which meet the safety, transport, and disposal regulations in force in France.

### 5.3 Experience

1. 2 m<sup>3</sup> of IERs have been embedded at Triton (anionic and cationic), in ten drums, which have been disposed of.

This campaign is being followed by the embedding of IERs from the D<sub>2</sub>O and H<sub>2</sub>O purification system of power reactor EL4 (250 MWth - 70 MWe), shut down in 1985.

## 6. New Generation of Mobile Unit

The epoxy process has been selected by EDF for embedding spent resins of French nuclear power plants with a new mobile unit.

This unit is intended to replace the units currently in service, as it offers epoxy characteristics which meet the latest French regulations.

The mobile unit is designed and constructed for embedding IER with various radioactivity characteristics (from less than 1 GBq/m<sup>3</sup> IER up to 52,000 GBq/m<sup>3</sup> IER).

The principal feature of this unit is adaptation of IER content so as to obtain a contact dose rate on the outer face of the concrete shell below 2 mSv.

Two types of concrete shell, in terms of dimensions and volume, are used.

IER content volume varies from 50 to 400 l. The thickness of steel/concrete between the embedded IER and the outer face of the shell forms a biological shield.

## Conclusions

Besides its applications to waste generated by operating plants (power reactors, research center, etc.), the epoxy process developed jointly by the CEA and TECHNICATOME has been used during various dismantling operations:

- fixing of contamination ,
- embedding of metallic waste during its packaging ,

- embedding of spent IERs, and
- barrier to limit tritium release.

The various applications demonstrate the qualities of radioelements containment by epoxy resins, and also the great flexibility in using this process.

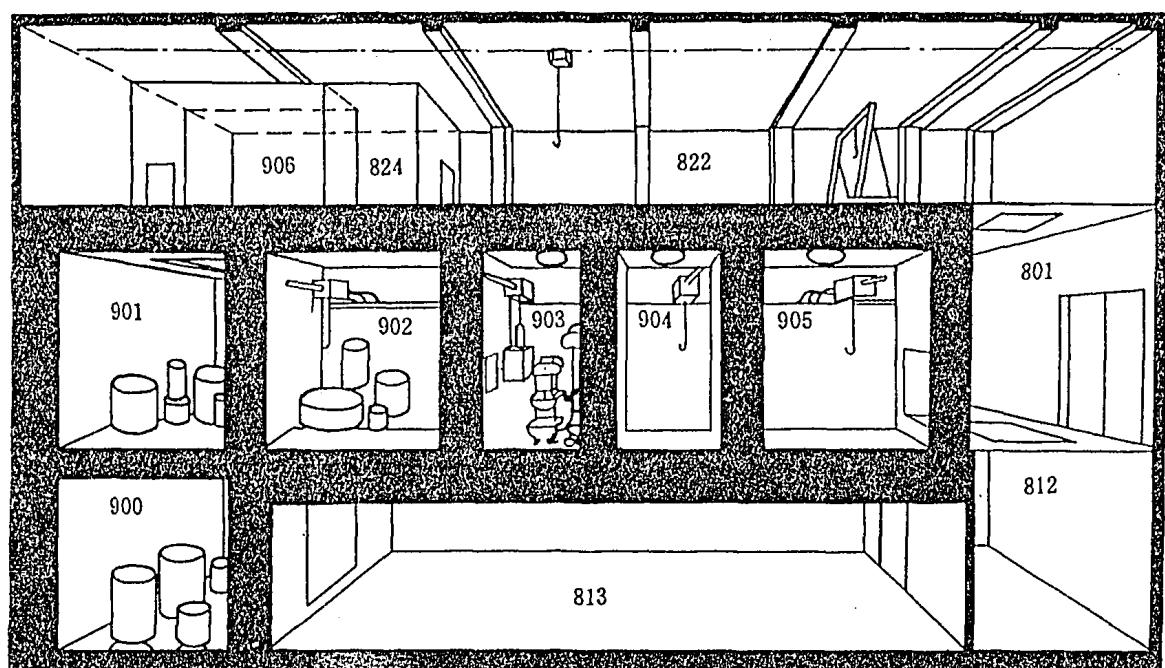


Fig. 1 ELAN 2B process cells

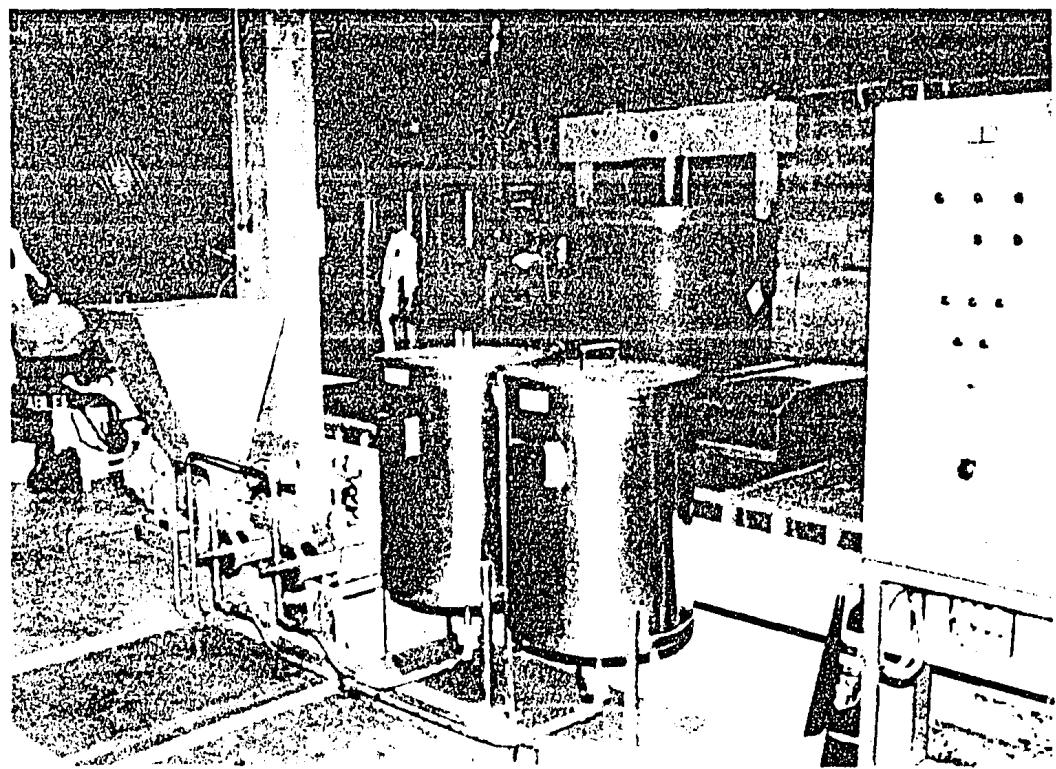


Fig. 2 ELAN 2B embedding unit

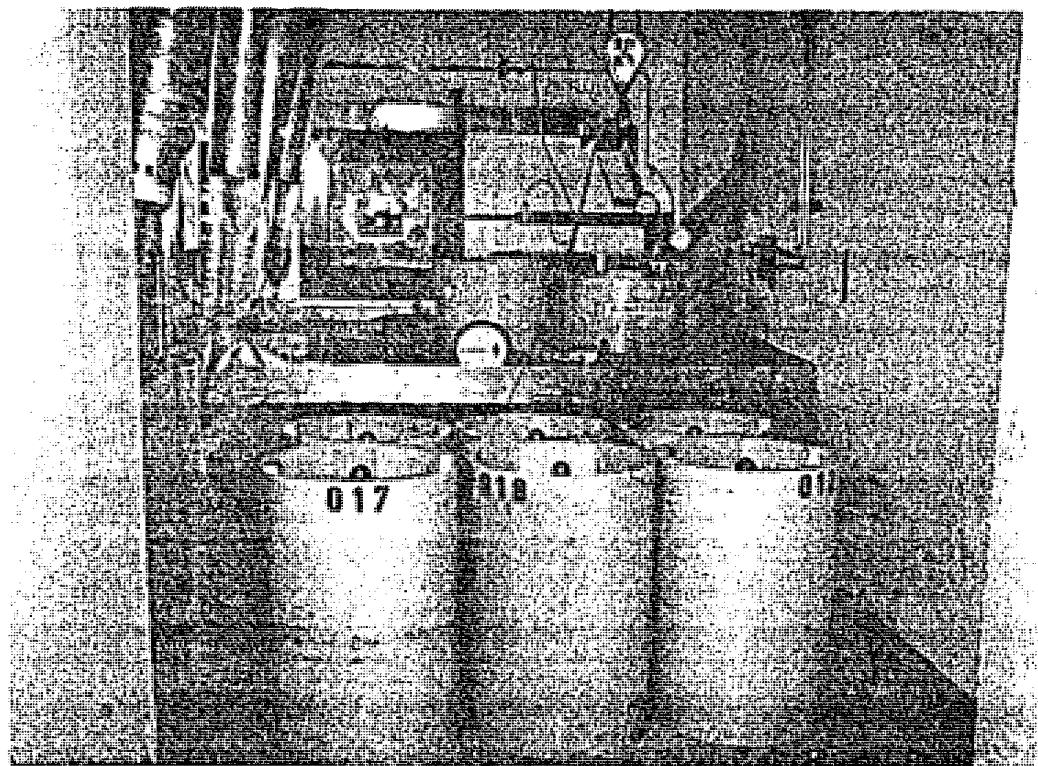


Fig. 3 ELAN 2B embedding unit

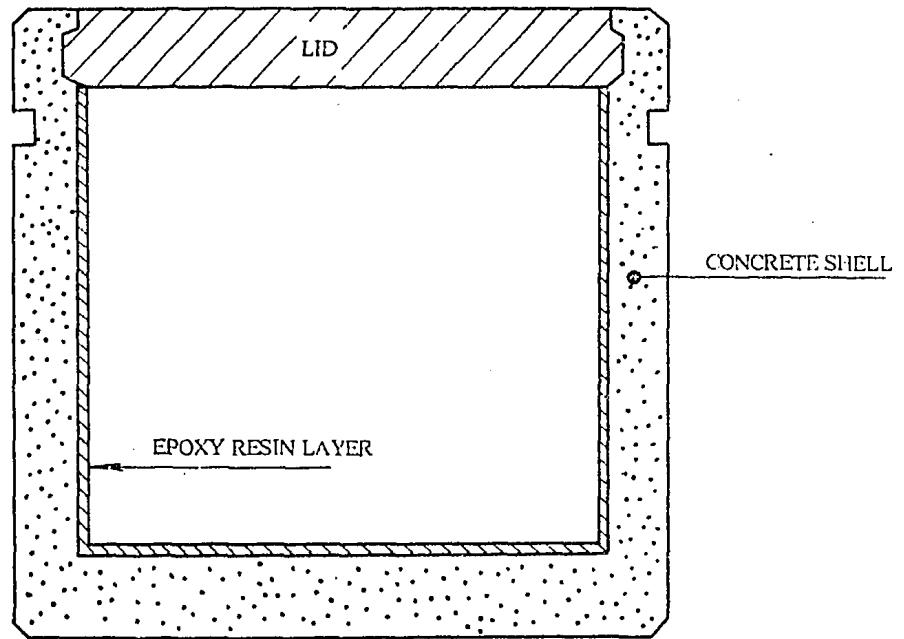


Fig. 4 EL3 concrete container

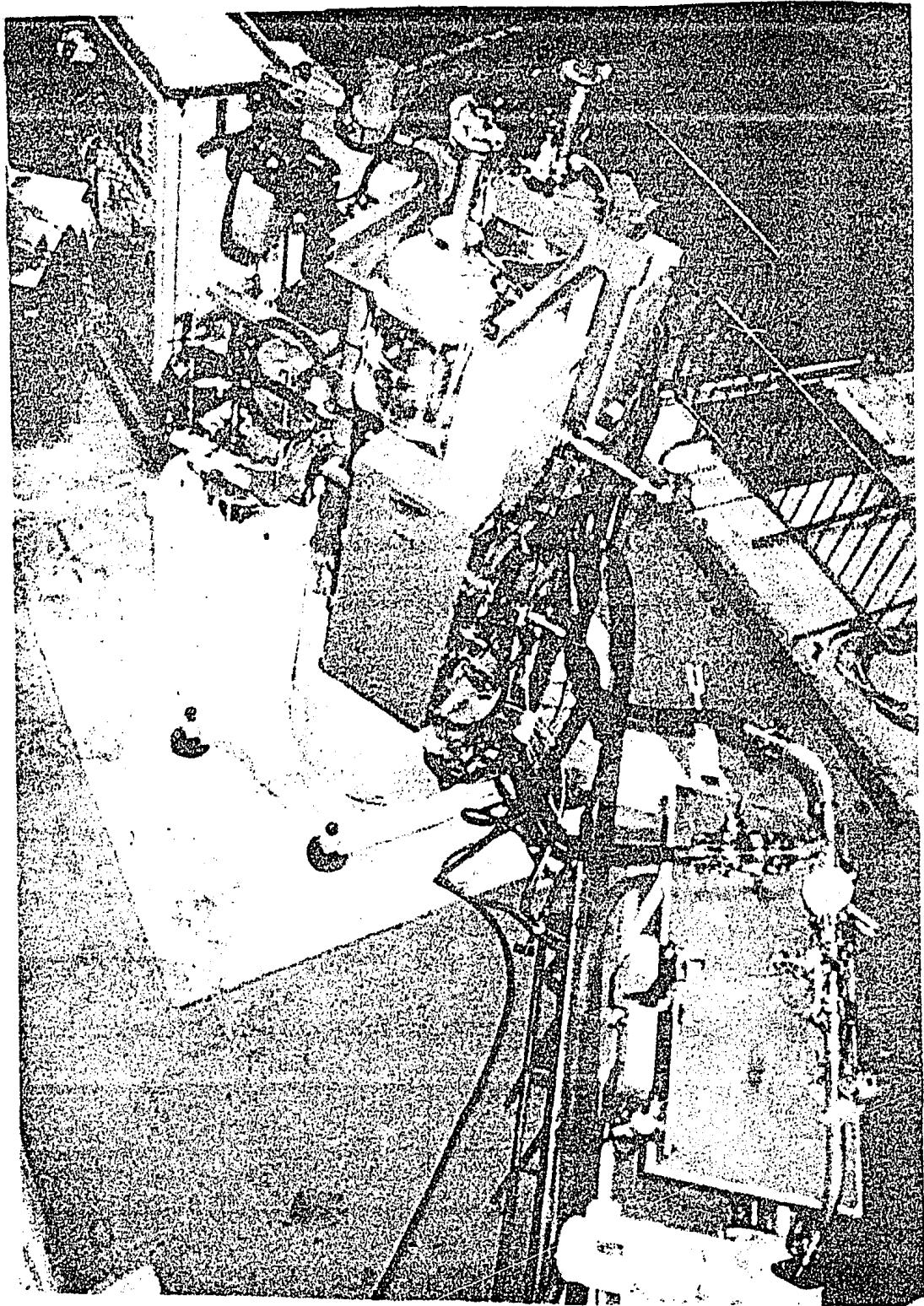


Fig. 5 SETH-200 Mobile Unit

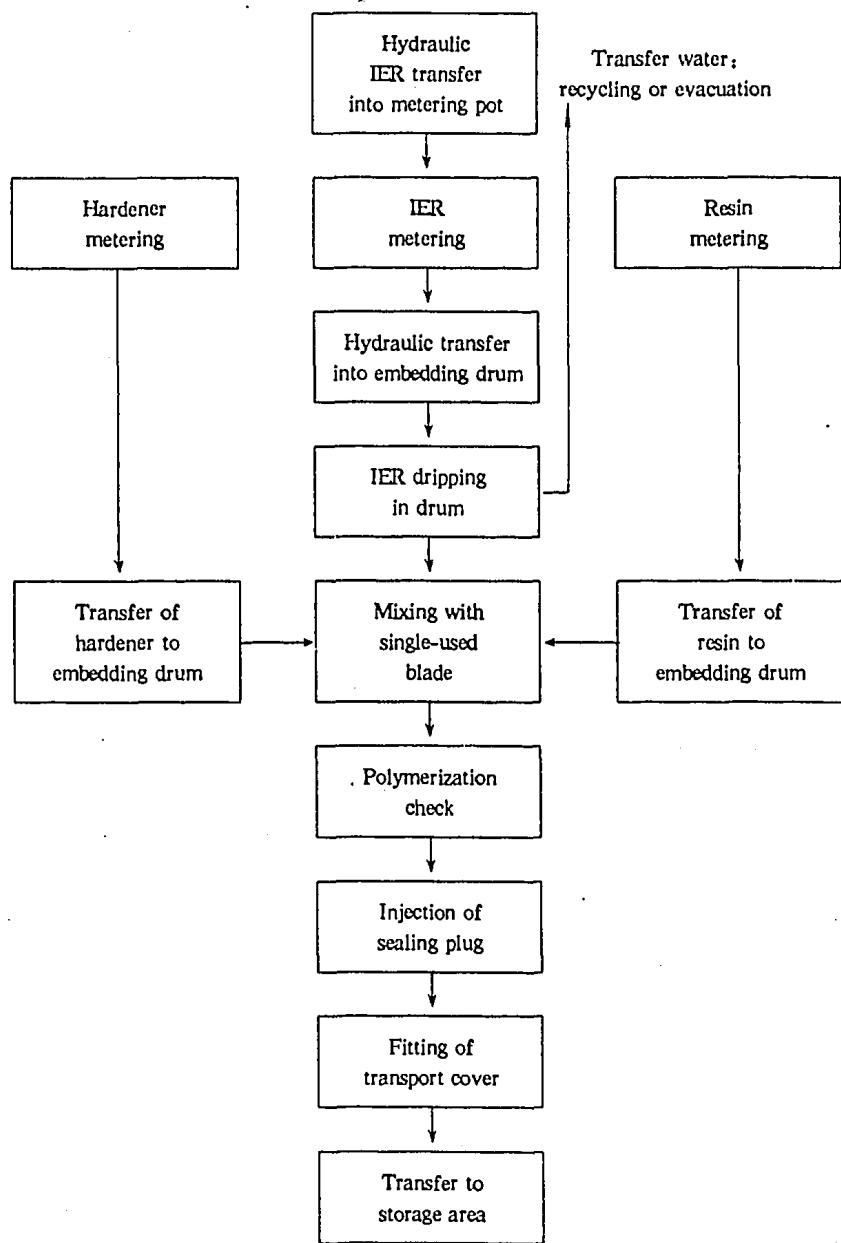


Fig. 6 Embedding Process in Technicatome Mobile Unit SETH-200

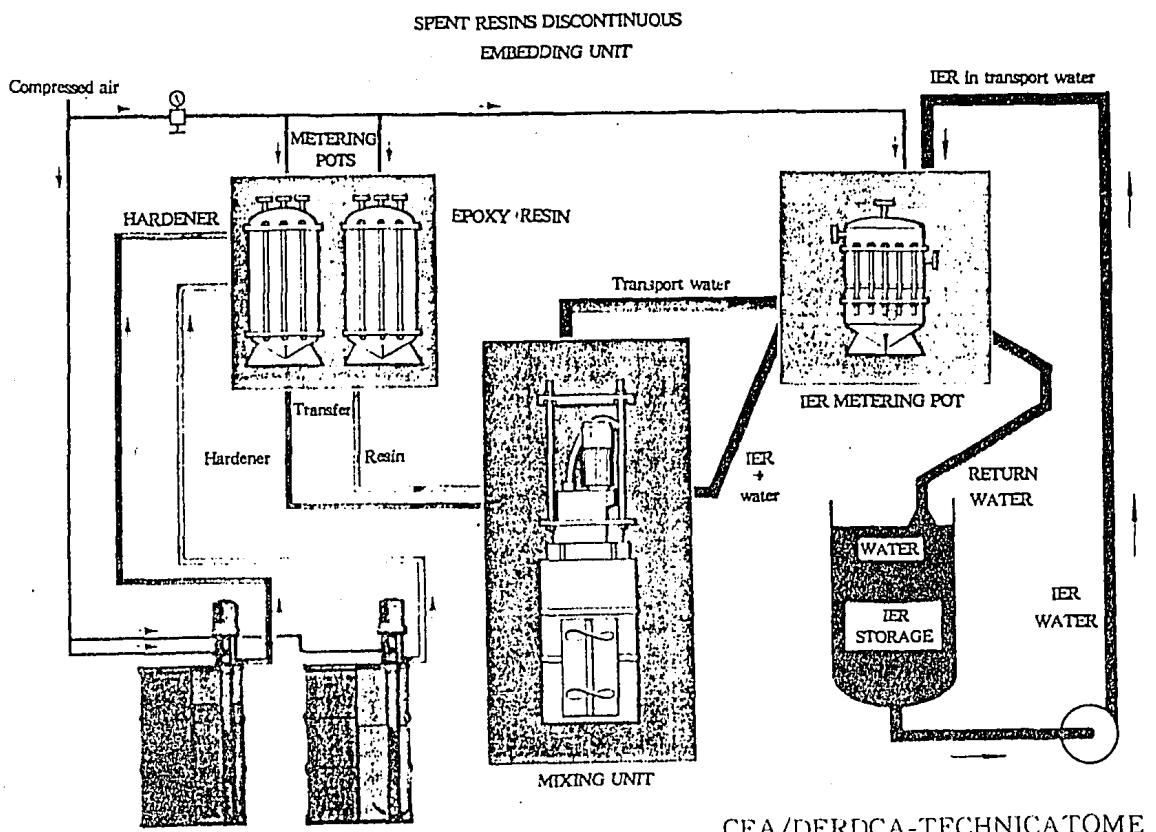


Fig. 7 SETH-200 flowsheet

### References

- [1] BUZONNIERE, A. AUGUSTIN, X. Embedding waste in thermosetting resins. In: CEA 75 Paris (France) Radioactive waste management decommissioning spent fuel storage V. 2 pp. 242-259.
- [2] LOURME, P. CHARAMATHIEU, A. Specific aspects of decommissioning. In: CEA 75 Paris (France) Radioactive waste management decommissioning spent fuel storage. V. 1 pp. 108-141.
- [3] JEANJACQUES, M. Decommissioning of nuclear plants, examples of application to workshops. In: CEA 75 Paris (France) Radioactive waste management decommissioning spent fuel storage. V. 1 pp. 142-171.
- [4] ROGER, J. Decommissioning of nuclear plants, examples of application to reactors. In: CEA 75 Paris (France) Radioactive waste management decommissioning spent fuel storage. V. 1 pp. 172-205.

# **BITUMEN AND CEMENT SOLIDIFICATIONS OF LL AND ML LIQUID RADWASTE**

## **THE SGN EXPERIENCE**

Eric TCHEMITCHEFF      Patrice ROUX  
SGN, FRANCE

### **1. Introduction**

The products from LL/ML liquid radwaste treatment — the corresponding operations are addressed in another paper — are evaporation concentrates, chemical sludge or ion exchange resins whose activity has been concentrated in a reduced volume but whose physicochemical characteristics are not consistent with final disposal requirements.

Conditioning is thus necessary to convert the waste into a solid and stable form able to meet the requirements set by competent Waste Management/Disposal Authorities as regards containment and mechanical resistance.

Relying on the extensive R&D work conducted by the CEA or our own laboratory teams on the one hand, and on a number of commercial applications on the other, SGN has soon acquired full industrial mastery of two conditioning processes, namely bituminization and concreting, that can cover the needs of the nuclear fuel cycle industry.

### **2. Bituminization**

As early as 1970, SGN was licensed by the CEA for the bituminization of LL and ML radwaste, either with extruders or thin-film evaporators.

About 15 systems were thus commissioned for industrial service throughout the world, providing extensive feedback of operating experience in the bituminization of a wide variety of waste: chemical coprecipitation sludge; PWR and BWR and research centers concentrates; resins from power plants and reprocessing plants, decontamination waste potentially including complexing agents, etc.

This presentation is focused on the thin-film evaporator technology and the experience gained in the field of the NPPs and research centers radwaste conditioning.

Laboratory tests on continuous bituminization process began in 1963. In parallel to extruder systems, the CEA developed a thin-film evaporator bituminization system which combines, when embedding liquid waste in straight distillation bitumen, a good quality of the final product and the following advantages:

- short residential time of the product in the evaporator, preventing its thermal degradation,
- low cost of investment,
- easy maintenance and operation, and
- high evaporation rates, up to 350 l/h of water evaporated.

The one-step volume reduction and bitumen solidification concept is a physical process that provides reliability and economy. A thin-film evaporator, operating at a waste product

temperature of about 160°C, is used to evaporate all free water from the waste influents. The remaining solids are homogeneously dispersed in the bitumen matrix while inside the evaporator.

The prepared waste is continuously fed at a controlled rate to the evaporator. Molten bitumen is simultaneously metered into the evaporator through a second feed nozzle. The evaporator is heated by means of a synthetic heating fluid circulated through an external jacket. As both the radwaste and bitumen are fed into the evaporator, the rotor blades spread the two streams into a thin, turbulent film against the heated internal surface. The action of the rotor blades and the force of gravity creates a spiral flow of the waste/bitumen mixture. As the waste flows downward through the evaporator, water is evaporated and the vapor flows counter-currently upward and out. The remaining radwaste mixture exits through the bottom of the evaporator into standard metallic drums. The evaporator discharge cone provides a buffer capacity and is fitted with a pouring valve in order to enable changing the drums without stopping evaporator feedings. Upon cooling, the waste/bitumen mixture solidifies into a free-standing, monolithic, water-free solid acceptable for storage or disposal.

The vapor leaving the evaporator is condensed in a shell and tube condenser and flows into the distillate collection tank. When this tank is filled, the distillate is pumped through a series of filters. The monitored cleaned distillate is pumped to the plant liquid waste system.

The whole system is fully automated and usually operated from a centralized control room.

In parallel with the commissioning of numerous facilities (see the reference list attached), large-scale R&D and technological development programs were carried out to continuously improve the existing technology and determine the optimum conditions for preparation of the encapsulated materials. Their goal was to meet the increasingly stringent requirements imposed by Safety Authorities for the operating safety of the facilities and for the long-term safe storage of the conditioned waste.

With the support of the CEA, SGN focused on three points:

- the development of pretreatments and operating conditions enabling safer preparation of the encapsulated materials,
- the development of pretreatments and formulations to obtain encapsulated materials with excellent long-term properties, and
- the design of systems enabling preservation of containment integrity throughout the process, particularly during preparation of the encapsulated materials and the pouring of these materials to fill drums.

As far as the first point is concerned, fire hazards were considered using a comprehensive approach at the conceptual design stage and during operation of the bituminization facilities, where there are basically three combustible elements in the process: the bitumen, the heating fluid, and the bituminized product. The bituminization process units are well instrumented and interlocked to prevent fire initiation, to eliminate the spread of a fire, should one originate from an outside source, to prevent release of radioactivity to the environment beyond permissible limits, and to protect equipment from possible damage. Redundant instruments and interlocks are provided for control points which, when exceeded, could become a hazard, namely the bitumen and thermal fluid distribution systems, and the part of the process where the bituminized product is prepared.

Even if this preparation of bituminized product occurs while permanently monitoring

temperatures, complementary measurements must be taken. More particularly, considerable attention was paid to the knowledge and concentration of chemicals which may be reactive or have high reducing strength to ensure safe operation of the bituminization process.

The extensive R&D work conducted by the CEA since the mid-1960s on waste of different compositions and forms helped to determine waste types that are consistent with the safe preparation of very-high-quality bituminized product. This also enabled acceptable composition ranges to be specified for various chemical species liable to occur, together with the best pretreatment method to be implemented, if necessary, against any risk of exothermal reaction that might incur fire hazards or unacceptable gaseous releases.

Operating parameters could also be specified, namely flow rates and temperatures.

Fully mastered recommendations could therefore be established, the most significant ones being the following :

- limitation of the reducing power of the waste,
- adjustment of its pH value (neutral or slightly alkaline),
- limitation of its contents of ammonium nitrate, chlorates or perchlorates, complexing agents, ferric or aluminium chloride,
- salt content in the end-product (wt salts/bitumen ratio: 35 to 50), and
- continuous monitoring of the pouring temperature for encapsulated materials. In this connection, SGN and the CEA have developed several types of highly reliable components for measuring this essential parameter. The associated technologies are suited to requirements: direct viewing or fiber optic signal transmission infrared camera, thermal vision camera providing pouring maps, including the temperature of each product droplet.

Although the probability of a fire initiating is extremely low, adequate methods of detecting fires quickly and suppressing those that occur have been defined to limit their damage. The methods employed include Halon or CO<sub>2</sub> fire suppression systems, water deluge system positioned directly over the filled drums.

Concerning the second point, concurrent R&D work enabled the successful definition of encapsulated materials and operating parameters meeting the increasingly stringent requirements of radwaste storage authorities for the quality of the encapsulated materials. This design focused on mechanical properties and containment capacities of the encapsulated species to ensure the long-term safety of permanent storage facilities.

The most significant methods are:

- insolubilization of radionuclides such as cesium or strontium by chemical coprecipitation to improve leach resistance, and
- conversion of hygroscopic substances to insoluble compounds to control swelling and even to avoid destruction of the embedded product when immersed in water.

Finally, concerning the third point, i.e. to resolve the essential problem of ensuring containment integrity throughout radwaste conditioning, the facilities are first designed on the basis of conventional nuclear engineering concepts: vented leaktight tanks connected to a system for the treatment of off-gas before release into the environment. The equipment used for bituminization (extruder and thin-film evaporator) are also leak-tight and thus fully consistent with this approach. Technical refinements have been introduced for the sensitive point of the drum filling station to enable design of ventilation hood systems which ensure dynamic

containment using air suction and trapping of gases produced during pouring of the bitumen encapsulated materials, as well as prevention of any splashing by encapsulated materials droplets.

### 3. Cement Solidification

SGN's involvement in radwaste cement solidification started up about 15 years ago with the design and commissioning of systems based on the conventional technology of the in-drum mixing.

Concrete solidification technology has been applied to different types of waste; borated concentrates, ion-exchange resins, etc. for many years throughout the world at reactors and research centers. Changing specifications for storage of radioactive waste have, however, confronted the operators of such facilities with two types of very different problems;

- binder/waste interactions, mainly when resins were concreted, and
- process application difficulties (unsuitable mixing methods, inaccurate metering, etc.).

With the support of EDF and COGEMA, SGN has been performing in-depth research on concrete solidification of borated concentrates and ion exchange resins generated by reactors or reprocessing plants, since 1983. At the same time it has been developing application technologies adapted to nuclear service conditions and stringent finished product quality requirements.

Processes developed by SGN from this research are based on the principle of a single, rapid pretreatment using available, inexpensive reagents associated, when resins are concreted, with a suitable hydraulic binder which evolve little heat while setting. Pretreatment can be adapted to various possible cases. Chemical reactions are controlled, which prevents exchange with cement calcium ions and, where necessary, also treats ions that interact with the binder (borates or ammonia).

The technology developed simultaneously by SGN (which meant refining both process and technology) is based on the use of a highly efficient, stainless steel batch mixer. Its design was refined on the basis of more than three years of pilot plant experience and the operation of a commercial system for seven years at the Valduc production center of the French Atomic Energy Commission (CEA).

The mixer is mainly characterized by the particular shape of its stirrer, which is located at the tank bottom. The stirrer transmits three successive movements to the product:

- slow horizontal displacement,
- rapid rising, and
- helical fall in vortex formed by the concave surfaces of the mixing paddle.

High efficiency and high encapsulating performances are achieved with this low-speed, yet high-energy mixing system systematically mounted on weighing scales for optimum process control. Mixing time is very short, i.e. less than 5 minutes.

The shape of the mixer, its construction material, and its simple design ensure that almost none of the products is retained in the tank. In addition, its specially designed rinse system limits daily waste water production for a 250-liter mixer to 10 liters (with little solids content).

The mixer is an air-tight tank and can contain liquids without any leakage, a very advantageous feature which enables in-mixer pretreatment operations prior to any feeding of solid materials. It

also prevents any splashing or external contamination.

Liquid wastes to be processed are due to be sampled in the storage tanks of the facility. Their chemical composition is well known and will be used to define the right formula for the concrete and to adjust the required amounts of dry additives such as cement and sand.

Pretreatment could be provided for specific effluents to make them compatible with their embedding into concrete or to improve the quality of the end product. This function would be performed directly inside the mixer or upstream from it in additional components.

In every case the liquid wastes are pumped directly into the mixer, whose stirrer has been put into operation. Then the cement and the solid additives are successively fed into the mixer by a screw conveyor. Each quantity of liquid or solid product needed to prepare a batch is automatically weighed as the mixer is set on a weight scale.

The resulting homogeneous mix of radwaste and cement is discharged by gravity directly into a shipping container, where it hardens into a monolithic solid. If necessary, according to the volume of the container to be filled, several batches are prepared successively.

It should be mentioned that inactive liquid grouts can be prepared, when replacing liquid wastes by water. Such inactive grouts can be used to immobilize filter cartridges or miscellaneous solid waste which have been introduced into a container before transfer to the concreting system.

This fully mastered technology aiming to enhance safe operation of its cement solidification process allowed SGN to face the very varied requirements of the different operators, such as alpha waste conditioning (CEA Research Center), medium active resins ( $300 \text{ Ci/m}^3$ ) concreting for EDF and COGEMA and pulverulent waste embedding such as pyrolysis ashes for COGEMA.

As for bituminization, this mastery allowed to extend this technology to the mobile units concept taking advantage of the stainless steel leak-tight mixer and specifically designed drum filling stations where the mixer is connected to the drum through a gasketed hood or a glove box with a plastic connector.

A first mobile unit was ordered by EDF for cement solidification of resins from the PWR secondary system and enables treatment of resins from eighteen 1,300 MWe PWR units, i.e. about  $180 \text{ m}^3$  of resins with a volumetric encapsulation ratio of 40 to 75%. The capacity of this plant is therefore about  $1.5 \text{ m}^3/\text{day}$  for one shift. The mobile unit is made up of three palletizable modules, which are transported between sites on a standard flatbed road trailer:

- a pretreatment module, which comprises agitated tanks linked to a gas processing unit (for removal of ammonia):

- empty weight:  $\approx 1.5 \text{ t}$
- overall dimensions: length: 3 m  
width: 2.2 m  
height: 3 m

- a dry load feed module, which comprises a hopper and a conveyor screw:

- empty weight:  $\approx 1.5 \text{ t}$
- overall dimensions: length: 2.5 m  
width: 2.2 m  
height: 3.5 m

- a process module, which contains the 250 l mixer, the ion exchange resins metering pot, the pouring station, the pouring drum handling equipment, and the gas processing facility:

- empty weight:  $\approx$  4.5 t
- overall dimensions: length: 3 m  
width: 2.2 m  
height: 3.45 m

This plant enables production of encapsulated material approved by storage authorities, regardless the type of resin involved (cation, anion or combined cation/anion, with variable saturation rates). It fills drums to optimum level ( $>95\%$ ), guarantees a reproducible quality level, permits easy, flexible operation and ensures minimum maintenance.

In addition to this project distinguished more by the chemical properties than the radioactivity of the waste, SGN has designed for EDF and foreign utilities mobile concrete solidification systems capable of processing radwaste that is substantially more radioactive, e.g. evaporator concentrates and reactor coolant system resins. As for the previously described system, the key component in these systems is the mixer. By exploiting one of its primary characteristics, the total leaktightness of its tank, the mixer can contain liquids without any leakage, a highly advantageous feature which allows in-mixer pretreatment operations prior to any feeding of solid material; it also prevents any splashing or external contamination.

These systems are also of modular design. They differ from the previously described system, however, because they require radiological shielding in the form of painted steel panels fastened to the metal support structures.

The modular design employed meets specific plant operator requirements and all the modules are usually shipped from one site to another on two standard-gauge road transport flatbed trailers. If necessary, the modules can be installed in an existing or specially built concrete structure, but the concreting system will still be fully tested in the assembly workshop and delivered practically ready for use.

**Bitumen Solidification Systems with Thin-Film Evaporator**  
**SGN Reference List**

FACILITY	OPERATOR	TYPE OF FACILITY	WASTE TYPE	EVAPORATOR CAPACITY (WATER EVAPORATED)	BITUMEN SYSTEM STARTUP DATE	DRUMS OF SOLIDIFIED WASTES
Barsebeck Nuclear Power Station	Sydkraft	Two 590 MWe BWRs	Bead resin, powdered resins, (sodium sulfate)	100 l/h	1975	6,000
Mihama Nuclear Power Station	Kansai Electric Power Co.	Three PWRs 320, 470, and 780 MWe	Boric acid, laundry, decon, chemical	200 l/h	1978	No Record
Tsuruga Nuclear Power Station	Japan Atomic Power Co.	375 MWe BWR	Sodium sulfate, decon chemical laundry	200 l/h	1977	No Record
Advanced Thermal Reactor, Tsuruga	Power Reactor and Nuclear Fuel Development	200 MWe LWC/IWR	Equip. and floor drains	200 l/h	1977	No Record
Valduc Center	CEA	Military Weapons	Sludges, concentrates	50 l/h	1970 to 1982	1,000
Cadarache Nuclear Research Center	CEA	Research & Development	Various	50 l/h	• 1971 to 1979 • 1986	No Record
Saclay Nuclear Research Center	CEA	Research & Development	Decon, EDTA, phosphates, nitrates, chlorides, ammonia, acids, bases	50 l/h	1975	3,200
Monts d'Arree Nuclear Power Station	EDF	70 MWe GCHWR	Sodium nitrate, phosphates	50 l/h	1981 to 1983	850
Oconee Nuclear Power Station	Duke Power Co.	Three 846 MWe PWRs	Boric acid, bead and powdered resins, decon	350 l/h	1983	—
Daejon Nuclear Research Center	KAERI	Research & Development	Concentrates, resins, sludges	50 l/h	1987	No Record
Clinton Nuclear Power Station	Illinois Power Co.	BWR 930 MWe	Sodium sulfate, bead and powdered resins*	200 l/h	1986 **	1500
Palo Verde Nuclear Power Station	Arizona Public Service Co.	Three 1221 MWe PWRs	Boric acid, sodium sulfate, bead resins*	200 l/h	1987 **	
North Anna Nuclear Power Station	Virginia Power Co.	Two 781 MWe PWRs	Boric acid, resins	200 l/g	1990	—
Sarry Nuclear Power Station	Virginia Power Co.	Two 915 MWe PWRs	Boric acid, resins	200 l/h	1991	—
Temelin Nuclear Power Station	CEZ	Two 1000 MWe PWRs	Boric acid, resins sludges	200 l/h	Under design	—

\* Processed by the same mobile unit

\*\* Start-up of the bituminization campaign

# LARGE VOLUME GROUTING SOLIDIFICATION AND DISPOSAL OF ILLW IN CHINA

SUN Mingsheng

Beijing Institute of Nuclear Engineering, China

## ABSTRACT

A large volume grouting solidification and disposal process for treating intermediate-level liquid waste (ILLW) in near-surface has been developed in China. The ILLW comes from the Lanzhou Reprocessing Plant. This process has been demonstrated by the cold test with simulated waste, and this project is now under implementation on the basis of design and construction.

### 1. Introduction

In recent years, R&D of radwaste management in China have been focused on the fields of solidification and disposal. Cementation process has been selected for the solidification of ILLW from nuclear power plants and reprocessing plants; and near surface burial has been adopted to dispose of low- and intermediate-level radwaste (L/ILW).

The Lanzhou Reprocessing Plant is the first of the kind in China, which is located in the Gobi Desert of Gansu Province with low population density and dry climate. Since the plant was put into operation, a great volume of ILLW has been accumulated and stored in carbon steel tanks. Although the large underground tanks can provide a safe interim storage condition, the lifetime is limited in light of long-term safe isolation of wastes. Because the ILLW has been stored there for more than 20 years, the waste should be solidified as early as possible.

The geological survey and evaluation of the disposal site was completed in 1980s, the proposed site is only 2.5 km away from the plant, where the groundwater level is about 40 m below the surface, and the geologic formation is very stable. In consideration of the above favourable conditions and reducing the costs of solidification, packaging, transportation, and disposal of the waste, an option of large volume grouting solidification and disposal process has been adopted. By this way, the radiation exposure to the operating and maintenance personnel can be kept "as low as reasonably achievable (ALARA)".

### 2. Characteristics of ILLW

The present ILLW of the reprocessing plant is classified into two categories of concentrated liquid radwastes, one contains sodium meta-aluminate generated from the dissolution of fuel elements of aluminum alloy cladding, its chemical composition and radioactivity concentration are as below:

Chemical composition	NaNO <sub>3</sub>	280 g/l
	Na <sub>2</sub> CO <sub>3</sub>	40 g/l
	NaOH	80 g/l
	NaAlO <sub>2</sub>	100 g/l

Radioactivity concentration	$^{90}\text{Sr}$	0. 056 GBq/l
	$^{137}\text{Cs}$	1. 32 GBq/l
	$^{106}\text{Ru} - ^{106}\text{Rh}$	0. 28 GBq/l
	$\Sigma\alpha$	26 kBq/l

The other is the neutralized raffinate concentrates coming from the second uranium cycle and the plutonium purification and extraction cycle, its chemical composition and radioactivity concentration are as below:

Chemical composition	$\text{NaNO}_3$	330 g/l
	$\text{Na}_2\text{CO}_3$	50 g/l
	$\text{NaOH}$	20 g/l
Radioactivity concentration	$^{90}\text{Sr}$	0. 16 GBq/l
	$^{137}\text{Cs}$	4. 25 GBq/l
	$^{106}\text{Ru} - ^{106}\text{Rh}$	0. 58 GBq/l
	$\Sigma\alpha$	67. 5 kBq/l
	Slurry $\Sigma\alpha$	51. 8 MBq/l

### 3. Formulation Research and Cold Test for Demonstration

Studies have been conducted to develop formulations for reducing the scope of the mixture of simulated liquid waste with Portland cement. Various grout formulations with different mix ratios of cement to simulated waste have been tested under laboratory conditions, and the chemical and rheological properties of the formulation, analyzed. Those formulations which make the waste-cement grout suitable for casting and disposal can be accepted. Several types of additives have been tested to improve grout flowing and satisfy time requirement for initial setting. Studies show that only when a DH<sub>4</sub>A type additive (0. 3% of cement by weight), i. e. a highly effective dehydrating agent, is added, can the simulated waste-cement grout have satisfactory performance. The compressive strength of the two samples of the aged 570 d reaches 16. 6 MPa and 11. 8 MPa respectively; and the temperature rise reaches 76 °C and 57 °C respectively. The volume self-expansion coefficient of the aged 90d is  $730 \times 10^{-6}$ , and the rates of leachability of the two kinds of simulated grout wastes for  $^{134}\text{Cs}$  and  $^{85}\text{Sr}$  are as below:  $R_{42d}^{134}\text{Cs} < 1. 7 \times 10^{-3} \text{ cm/d}$ ,  $R_{42d}^{85}\text{Sr} < 5 \times 10^{-4} \text{ cm/d}$ ; and  $R_{42d}^{134}\text{Cs} < 3. 7 \times 10^{-3} \text{ cm/d}$ ,  $R_{42d}^{85}\text{Sr} < 1 \times 10^{-3} \text{ cm/d}$  respectively.

On the basis of formulation research, a pilot cold test for demonstration was carried out in 1986. In this experiment, the simulated ILLW and cement were fed into a vertical mixer with a double-impellerstirrer, the cement paste flowed out of the mixer and was casted into an underground concrete vault with a size of  $4. 2 \times 4. 2 \times 3. 5\text{m}$ . This is a continuous casting process with an output of  $10 \text{ m}^3$  cement paste per hour and the whole process was quite satisfactory, safe and reliable. The temperature of the cement solidified waste of simulated waste reached the maximum value, 119 °C, 5 days after casting and then it dropped to 35 °C, 60 days after casting. Its properties were as good as expected.

### 4. Project Design and Construction

The basic design for this project has been completed. The project consists of:

cement system,  
 ILLW collection and transfer system,  
 ILLW feed system, and  
 grouting disposal system.

Cement system and ILLW collection and transfer system are located in the reprocessing plant area, while ILLW feed system and grouting disposal system are arranged at the disposal site. ILLW is pumped from the collection and transfer system into the feed system, then into the mixer of the grouting disposal system. All the concrete vaults are placed underground, and the mixer sits at the top of the vault. The waste cement paste flows into the vaults by gravity, and the well-mixed grout stays in the mixer for about 8 minutes. A vault with a size of  $8 \times 8 \times 6$  m will solidify about  $230 \text{ m}^3$  of ILLW. The waste cement paste casting for a vault will take about 26 hours continuously. Several days later, an additional layer of clean cement paste will be cast on the surface of the solidified waste.

Every twelve vaults form a unit, and the number of units to be constructed depends on needs. This ongoing project is also expected to realize a unique disposal system to ensure the long-term protection of the public and the environment. Natural analogues are used to enhance the defensibility of the design features. The diagram of the grouting disposal system is shown in Fig. 1. The design features considered involves the isolation barriers for the following requirements:

- A Reinforced concrete vault with a structural cover,
- A multilayer protective barrier for reducing the amount of infiltrating water to reach the waste block and for preventing bio-intrusion, and
- A clay layer surrounding the vault to retard the release of radionuclides from the disposal system.

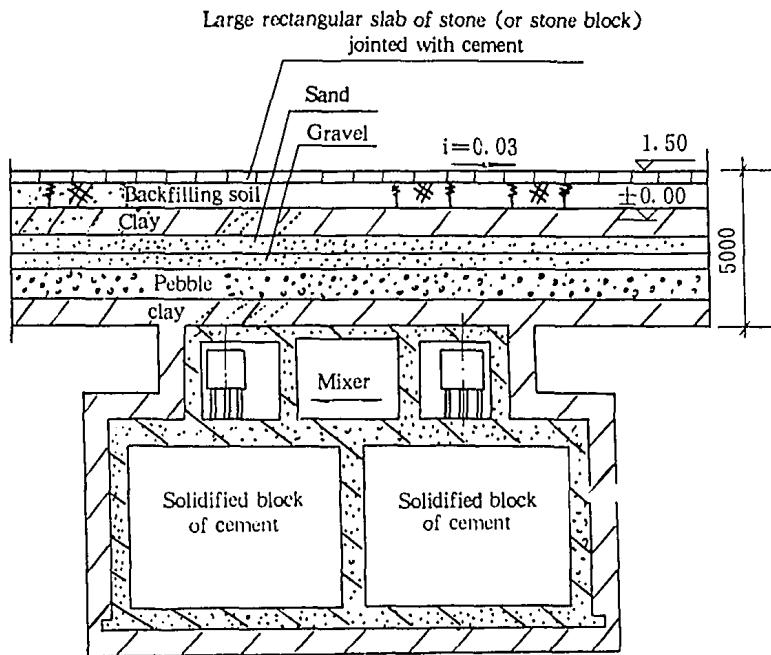


Fig. 1

# STUDY ON DEWATERING OF CHEMICAL SLUDGE BY FREEZE-THAW PROCESS

XU Shikun LIU Pin

China Institute of Atomic Energy

## 1. Introduction

Sludge is produced in treating industrial waste water, civil waste water, and radioactive waste water by flocculation. The sludge produced from treating radioactive waste water contains radioactivity. The treatment of radioactive sludge is different from that of non-radioactive sludge. One of the most commonly used methods is cementation, and the cement-solidified product is finally disposed of. The solid content in the low-level radioactive sludge is 1~2% initially, and will be 6~10% after long-term storage. This shows that there is a large amount of water in the low-level radioactive sludge. The volume of the sludge will be greatly reduced after dewatering. For the final disposal, the volume-reduction of this product has a high benefit in economy.

The methods to be used for sludge dewatering are mainly flocculation, filtration (natural filtration, pressure filtration, and vacuum filtration), and centrifugal separation. Since low-level sludge is a suspended colloid, not easy of dewatering. Therefore, the fast and large-scale dewatering of such sludge can not be achieved by the above methods. The freeze-thaw plus centrifugal separation and the freeze-thaw plus vacuum filtration have been developed for effective dewatering the unpretreated sludge separately at the Atomic Energy Research Establishment (AERE), Harwell, United Kingdom and the Nuclear Research Centre, Mol, Belgium. Volume-reduction factor (Volume-reduction factor = Volume of original sludge / Volume of sludge after freeze-thaw process and subsidence) of these two kinds of methods can be very high. The physical properties of the freeze-thawed sludge are completely different from those of the originals. The resulting solid is in a granular shape and settles well. The volume-reduction factor is about 5~7. If it is progressively separated through vacuum filtration and centrifugal separation, the volume-reduction factor or the solid content can be higher.<sup>[1]</sup>

In our laboratory, methods of immersing freeze and simulated two-step freeze have been studied for the elementary properties of simulated low-level sludge, the effect of freezing temperature, freezing time, and settling time on volume-reduction factor. Some parameters for design of freeze-thaw device are provided.

## 2. Experiment

### 2. 1 Experiment of freezing in a cryogenic bath (Type KF2 Cryogenic Bath)

The sketch of the device is shown in Fig. 1. The cryogenic bath was filled with 50% glycol solution as cooling medium. The plastics bottle (100 ml) was used as a container for freezing sludge and immersed in the coolant. The original sludge with 2% and 7% solid was prepared. Specimens were obtained after freezing, and the frozen sludge was then thawed in the atmosphere for 24 hours. After baking the specimen, the solid content was measured.

## **2. 2 Test of simulated two-step freeze**

A simulated freeze-thaw device consists of a small freezing tank, a small power pump, and a cryogenic bath as shown in Fig. 2. The parameters of freezing tank are:

Capacity : 0.8 liter  
External diameter of spiral coil :  $\phi 86$   
Internal diameter of spiral coil :  $\phi 70$   
External diameter of copper pipe :  $\phi 8$   
Internal diameter of copper pipe :  $\phi 5$   
Turns of spiral coil : 11  
Heat exchanging area :  $0.06292 \text{ m}^2$   
Solid content of original sludge : 2% and 7%.

The dewatering effects on sludge under different freezing temperatures and different time were measured.

## **2. 3 Pilot freeze-thaw test of 20 liter in capacity**

The process is shown schematically in Fig. 3. The capacity of freezing tank is 20 liter. The solid content of original sludge is 2%.

Heat transfer area of freezing tank is designed to  $0.8273 \text{ m}^3$ .

The pilot freeze-thaw facility with 20 liter in capacity needs cryogenic pump and other pumps suitable for proper flow, a compressor with sufficient power, proper proportion of stored freezing medium to working freezing medium. Under these conditions, the sludge can be ensured to be frozen completely within the required time .

## **3. Results and Discussions**

### **3. 1 Mechanism of flocculation by freeze-thaw process**

Crystallization of water originates from crystal nuclei and extends in branched shape. The simulated sludge contains calcium phosphate and ferric hydroxide. Part of the sludge is colloid, its particles are electrically charged. Fig. 4 gives the structure of a double electric layer of colloid particles. Colloid possesses special physical properties such as Brownian movement and electrophoresis, which are destroyed during freeze-thaw process. During freezing, the electrolyte accumulated around the sludge particles can destroy the electrical potential layer. Finally, the frozen and thawed sludge particles are aggregated under an action of the force between molecules to form granular particles which are easy to settle. Thus, the dewatering of sludge is realized.<sup>[2]</sup>

The ice point of sludge measured in the experiment is - 0.22 °C ; under the same conditions, the ice point of natural water, - 0.13 °C. The ice point of sludge is close to that of water.<sup>[3]</sup>

### **3. 2 Results of experiment in cryogenic bath**

The 7% sludge is frozen at different temperatures of - 6 °C , - 8 °C , and - 10 °C respectively. The results of freezing effects are shown in Fig. 5. The solid content of sludge rises rapidly to 20~22% after frozen for 4 hours at the temperatures of - 6 °C and - 8 °C , but only 18% at - 10 °C. This may be resulted from fast freezing at - 10 °C.

For 2% sludge, freezing effects at - 6°C, - 8°C, - 10 °C and - 12 °C are rather good, in which the best freezing effect is achieved at - 12 °C. But the result at - 14 °C is worse (see Fig. 6). From the figures, it can be seen that after the sludge being completely frozen, the solid content of the sludge will not be increased any more. The destruction of sludge properties occurs mainly during freezing of water. In case of fast freezing, the aim of dewatering can not be achieved.

From Figs. 5 and 6, it is found that, for 7% sludge and 2% sludge, the optimum freezing temperatures are - 8 °C and - 12 °C respectively. The optimum freezing time is 4 hours for both cases. It shows that solid content of sludge affects the freezing process.

It is observed from microscopy that the particle size ( $d$ ) of the frozen and thawed sludge will be decreased with the decrease of freezing temperatures, e.g.  $d(- 6 °C) > d(- 8 °C) > d(- 10 °C) > d(- 12 °C) > d(- 14 °C)$ . Therefore, when sludge is frozen slowly, the bigger volume-reduction factor can be obtained.<sup>[4]</sup>

### 3. 3 Results of simulated two-step freezing test

The conditions of this test are chosen according to the results as described above. The longest freezing time is 4 hours.

The 2% sludge is frozen at - 8 °C, - 12 °C, and - 15 °C respectively. After thorough freezing, the solid content of the frozen and thawed sludge reaches 18% ~ 20% at each given temperature. At the lower temperature, the solid content rises gradually; at the higher temperature, it rises steeply (see Fig. 7) after the sludge is frozen for 2 hours.

When the 2% sludge is frozen at - 8 °C, - 10 °C, - 12 °C, and - 16 °C, the volume-reduction factor is between 7 and 8, which shows a rising tendency. When the 7% sludge is frozen for 4 hours at - 8 °C, - 10 °C, - 14 °C, and - 16 °C, the volume-reduction factor is between 2 and 3, and will be descended slightly with descending of temperature, which is similar to the previous results. In order to achieve the aim of sludge dewatering, different freezing temperatures should be applied to different sludge solidcontents, see Fig. 8.

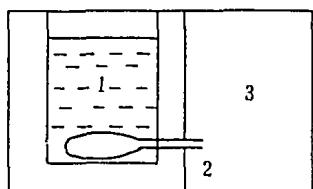
It is found from Figs. 5, 6, and 7 that the optimum freezing period is 4 hours; in shorter time, the sludge can not be frozen completely; if too long, the volume-reduction factor can not be gone up any more.

### 3. 4 Results of pilot freeze-thaw test of 20 liter in capacity

The sketch of process is shown in Fig. 3. The freezing temperatures are chosen at - 8 °C, - 11 °C, and - 15 °C. The solid content of original sludge is 2%. The results are shown in Fig. 9. It is found that - 11 °C is the optimum freezing temperature and 4 hours, the optimum freezing period. The volume-reduction factor can reach 6~7.

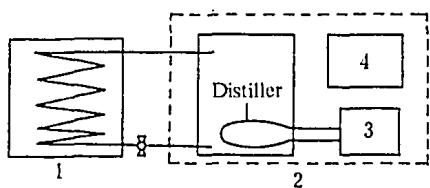
## Conclusions

The results of the above experiments show that the freeze-thaw process can be used to effectively dewater the sludge with the volume-reduction factor up to 6~7. The parameters obtained from these experiments are quite useful for engineering design of large-scale freeze-thaw process.



1. Cooling medium      2. Distiller  
3. Instruments and compressor

Fig. 1 Cryogenic bath ( $-30^{\circ}\text{C} \rightarrow 50^{\circ}\text{C}$ )



1. Freezing tank      2. Cryogenic bath ( $-30^{\circ}\text{C} \rightarrow 50^{\circ}\text{C}$ )  
3. Compressor      4. Controlling instruments

Fig. 2 Minitype two-step freezing device

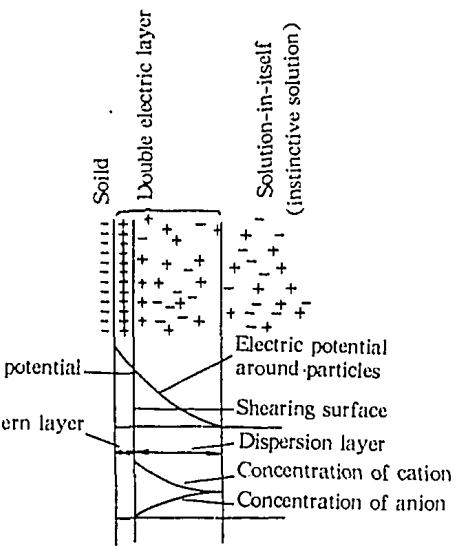
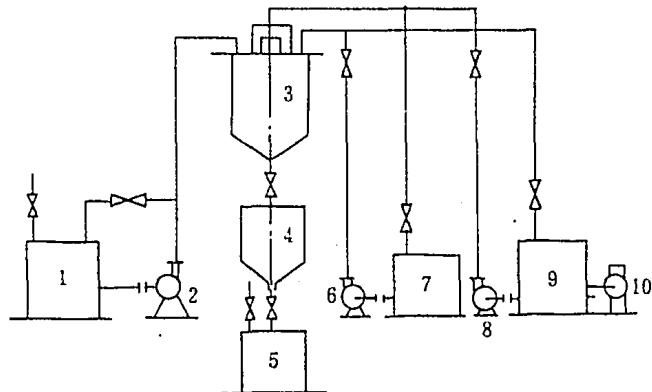


Fig. 4 Double electric layer



1. Sludge tank      2. Sludge pump      3. Freezing tank      4. Settling tank  
5. Sludge tank      6. Water pump      7. Thawing medium tank  
8. Water pump      9. Evaporator      10. Compressor system

Fig. 3 Technological process of 20 t capacity

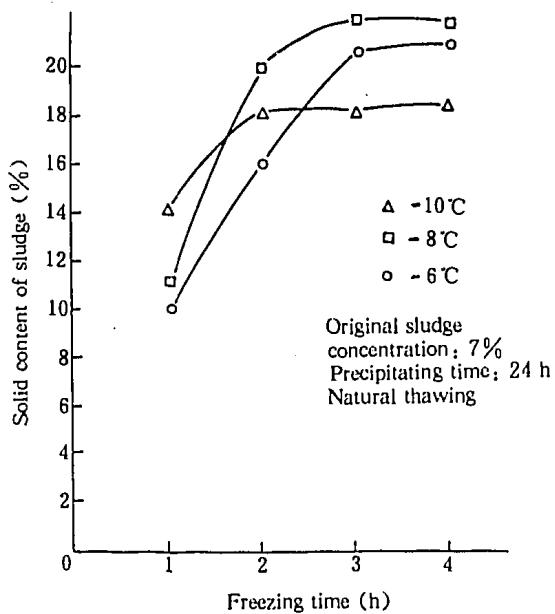


Fig. 5 Solid content of sludge versus freezing time in immersing freezing method

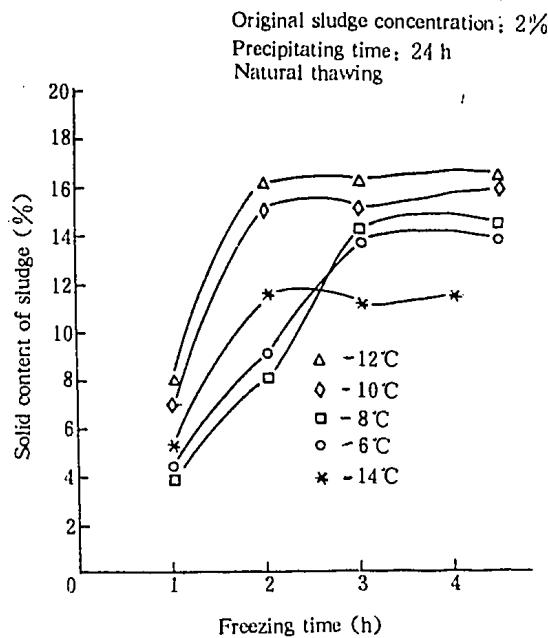


Fig. 6 Solid content of sludge versus freezing time in immersing freezing method

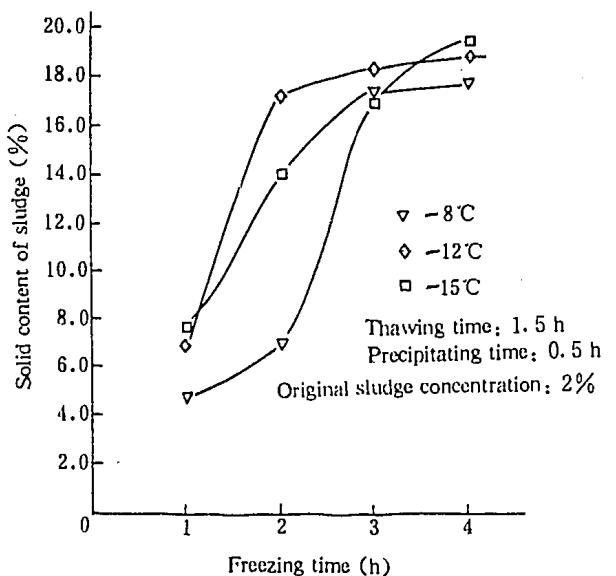


Fig. 7 Solid content of sludge against freezing time in a small-scale simulated two-step freezing test

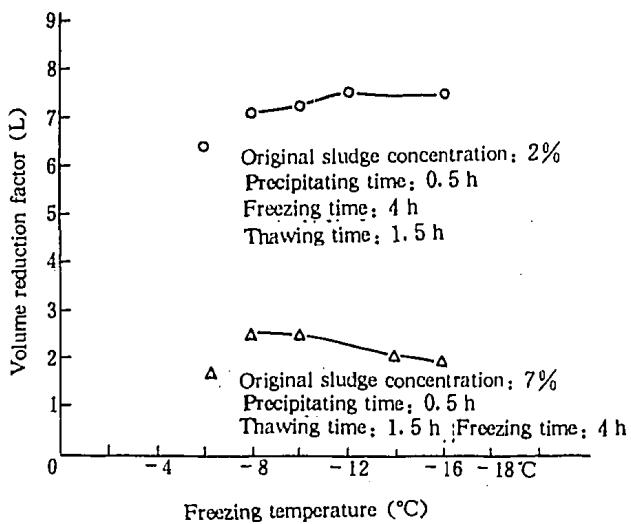


Fig. 8 Volume reduction factor against freezing temperature in a small-scale simulated two-step freezing test

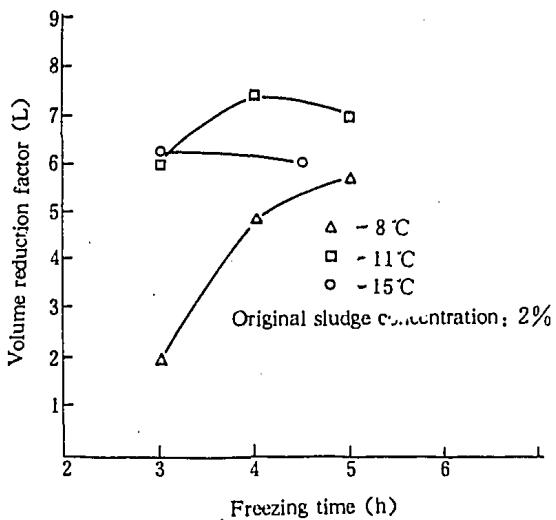


Fig. 9 Volume reduction factor versus freezing time in the test of 20 liter in capacity

## References

- [1] Treatment of Low- and Intermediate- Level Radioactive Wastes Concentrates, IAEA REPORT No. 80, 1968
- [2] Richard Akers, Flocculation, Pennvalt Limited, United Kingdom, 1975
- [3] XU Suzhen et al, Study on Cement Solidification of Radioactive Chemical Sludge, "Radiation Protection", Vol. 2, P96, 1986 (Chinese)
- [4] CHEN Zhuying, Study on Freeze-thaw Dewatering of Chemical Sludge, "Atomic Energy Science and Technology", Vol. 2, 1993 (to be published) (Chinese)

# SOLID RADWASTE PROCESSING AND CONDITIONING

## THE SGN EXPERIENCE

Daniel TUCOULAT      Eric TCHEMITSCHOFF  
SGN, FRANCE

### 1. Introduction

The operation of any nuclear installation generates solid waste. Nuclear installations in France include spent fuel reprocessing plants owned by COGEMA, nuclear power plants operated by the French Utility EDF, nuclear research centres of CEA (the French Atomic Energy Commission) but also research centres, hospitals, and universities — so called "small producers" — subject to centralized waste management. Solid wastes are of various types:

- so-called process waste such as structural waste of fuel elements in fuel reprocessing plants or filters installed on liquid or gaseous circuits in nuclear power plants, and
- so-called technological waste from maintenance operations. They include worn process equipment, clothes, paper, board, plastics, metal pieces, debris, gloves, protective clothing, etc.

This type of waste is the major issue addressed in this paper. These wastes display different levels of activity. Some of them arising from research centres or reprocessing plants even contain relatively significant quantities of alpha-bearing radionuclides.

As a general rule, solid waste conditioning takes place in a number of successive steps in order to:

- produce an ultimate waste package that satisfies the requirements set by Safety Authorities and the organizations in charge of final waste disposal in the concerned countries, and
- reduce as far as practicable the final volume of waste to be disposed of.

SGN has a full commercial mastery of the various technologies involved and knows how to combine them in the best possible way thanks to the experience it has gained from applications in France that can easily be transposed for foreign clients.

### 2. Processing Operations

Solid waste treatment operations include:

- sorting, shredding, cutting, and more generally all the operations preparatory to conditioning aimed to segregate the wastes into categories appropriate to the downstream conditioning and to standardize the wastes that are generally of very different sizes;
- counts intended to characterize activity of the solid waste in bags or drums, etc. ; and
- operations that intend to reduce the level of activity of  $\beta$ ,  $\gamma$  or  $\alpha$  bearing wastes before they are conditioned in order to simplify the technologies to be implemented.

Technological waste is most often packaged in bags, bins, drums or containers as close as possible to the source, but it may also be stored in bulk in big silos from where it must first be retrieved.

With the help of the operators of the COGEMA reprocessing plant, La Hague, SGN could develop a mobile enclosure concept designed to deal with problems of rehandling waste stored in silos which have no controlled servicing area in the upper part. These mobile enclosures are mounted on bogies, circulate on rails and contain the necessary bulk waste recovery means (grippers, ...) but also all mechanical equipment for waste "preconditioning" and placing in 200 l drums.

Mobile enclosures are designed to be airtight for maximum containment of the zones above the silos. To do this, a seal is provided with the silo to be emptied by means of a thick rubber strip and the building is ventilated by HEPA filters and exhausters.

Such waste recovery systems have successfully been operated at La Hague on very low  $\beta\gamma$  solid waste and are able to "produce" about forty 200 l drums of "preconditioned" waste per shift.

#### Solid waste sorting and preparation

The industrial control of integrated systems has been achieved, making it possible to design equipment in full consideration of the protection of the operators necessarily active at the workstations, to combine all or part of the equipment such as shears, metal detectors, precompactors, shredders like knife mills, and hydroextractors designed to separate the liquids when potentially present in the waste, in sealed glove boxes or shielded cells, with sealed connection on the drums removing the sorted wastes.

Bulk stored waste in particular must be sorted according to type and future conditioning. The methods and systems used include:

- manual sorting in glove boxes or shielded cells, using master-slave manipulators,
- line of vibratory screens with decreasing mesh,
- magnetic sorting machine, and
- eddy current machine for sorting by projection.

Typical crushers are equipped with:

- two shafts fitted with cutting disks,
- an inlet hopper fitted with a press arm which pushes the waste towards the disks and can even be used to fragment bulky waste such as filtration housings before they are crushed, and
- two electric motor drives.

In the same way, typical hydroextractors are equipped with a suspended bowl rotating at a high speed that allows continuous operation. The crushed wet wastes enter at the top and are discharged laterally by the centrifugal force into a cyclone separator, which slows them before sending them to a drum. The waste is recovered in a separate collecting drum.

Several contracts were thus awarded to SGN for the detail design of integrated solid waste treatment systems, namely for EDF for the treatment of LL metal waste arising from all the French nuclear power plants, for ONDRAF in Belgium and currently for ZWILAG in Switzerland (centralized conditioning facility for solid waste from the nuclear industry).

#### Decontamination

This operation is particularly concerned with metallic wastes and can be performed to:

- downgrade the waste for shallow-ground disposal,
- reduce the proportion of ML to LL waste, and

- facilitate subsequent conditioning operations by removing the labile activity and by lowering the overall level of activity.

With the feedback of the very large experience gained by the COGEMA operators, SGN can today apply proven methods both for in site operations and in dedicated facilities, efficiently integrating a set of methods and guaranteeing the containment and protection of the operators to remove labile or fixed contamination.

In addition to very "conventional" methods used for example in the AD1-BDH facility at La Hague, i. e. :

- chemical decontamination methods by soaking, combined when necessary with ultrasonic effects,
- electro-decontamination methods in stirred baths,
- high pressure jets whose efficiency can be enhanced by
  - increasing the water pressure, up to 300 bars,
  - adding chemical reagents,
  - using the thermal effect, and
  - adding an abrasive (usually angular or spherical - or glass beads depending on the surface texture).

SGN is involved in the development carried out by CEA of a number of innovating, very promising methods aimed at :

- significantly reducing the volume of secondary effluents,
- noticeably increasing efficiency, and
- efficiently solving problems linked to the recovery of plutonium from certain wastes rich in alpha emitters.

Some of these methods are listed below :

#### Spraying gels containing chemical reagents

The solution containing the reagents is sprayed after the addition of the gelling agent (organic, silica) up to viscosities ranging from 300 to 800 cps or more (thixotropic gel).

The gel remains in contact with the surface to be decontaminated for sufficient time for the reagents to act. The treated surface is then rinsed by a conventional method.

#### Electro-decontamination using a cathode tool

Electrolysis is carried out using a more or less sealed cathodic swab which is moved by the operator or by a robot in contact with the surface to be decontaminated. The electrolyte is applied only on the spots to be decontaminated, minimizing the volumes to be used and processed later.

#### Cryogenic abrasives

Abrasive spraying often generates large volumes of secondary waste that is difficult to recycle.

Another drawback of conventional abrasives is that particle impact on the surface to be cleaned tends to produce abrasive fragments appearing on the surface.

The use of a cryogenic abrasive, which is converted spontaneously to liquid or gas after spraying, helps to eliminate these problems. These techniques, which are the latest and the most promising, have been the subject of patents filed by the CEA.

- Ice

Water is injected through a distributor perforated with calibrated holes, and the drops, partly frozen in gaseous nitrogen, are then solidified in liquid nitrogen.

The beads (0.5 to 2 mm) are sprayed by an ice gun using nitrogen or dry air as a carrier gas.

- Dry ice

Pellets are produced (length 3 to 6 mm, dia. 3 mm) by expanding liquid CO<sub>2</sub> and the dry ice produced is compressed and extruded in a pelletizer.

The pellets are sent after screening to a batcher which feeds a spray system.

This system is in industrial use today.

Silver electrolytic decontamination method

This method was developed by the CEA, in particular for the decontamination of solid waste containing plutonium and even for the recovery of this contaminant.

Based on the action of Ag<sup>2+</sup> ions electrogenerated in a nitric medium on plutonium oxide, this method can be applied:

- to metallic waste, 10<sup>4</sup> decontamination factors being easily reached since decontamination is enhanced by Ag<sup>2+</sup> ions that initiate a corrosion of the metallic waste surface and facilitate transfer of contaminants to the solutions,
- to plastic waste where high efficiency can be reached for little aged waste resisting oxidation by Ag<sup>2+</sup> ions, and
- to the treatment of ashes resulting from the incineration of waste highly contaminated with plutonium, and hence which are rich in PuO<sub>2</sub>.

SGN is carrying out design and erection of an industrial facility at the La Hague reprocessing plant, based on that process.

### 3. Volume Reduction Techniques

To optimize the filling of waste drums and to reduce the volume after filling, and hence the volume of the waste sent to disposal, many volume reduction techniques are employed. These are mechanical (cutting, crushing, and compaction) or thermal (incineration) methods.

Cutting

Cutting is a preparatory operation more than a volume reduction technique even though reducing the bulk factor also reduces the overall waste volume.

Conventional cutting techniques are:

- sawing,
- shearing, and
- plasma torch cutting implemented for instance to cut metallic process waste in vitrification facilities.

However, efforts were made to nuclearize and remotely operate conventional tools by installing them, according to the level of activity of the waste to be processed:

- in dedicated bays allowing contact work while satisfying all personnel safety requirements, and
- in sealed boxes fitted with gloves or manipulators, if necessary shielded for the most irradiating waste.

### Compaction

This technology is widely used by the different entities involved in the fuel cycle, a wide variety of forms, ranging from low power systems (50 t) acting mainly in precompaction, to multi-directional presses (2D and 3D) producing parallelepipedal waste packages up to 1,500 t and even 2,000 t supercompaction installations, both fixed and mobile, working on  $\beta\gamma$  wastes alone or alpha wastes.

Industrial experience is considerable, and the initial "commercial" units are reliable. This has led to the commissioning of nuclearized equipment with easy maintenance, containment, mechanical components transferred outside the contaminated zone, and prevention of the risk of contamination of the drive oil. This applies in particular to the 1500 t press used by COGEMA at La Hague to compact contaminated alpha waste, arriving in 200 l drums (AD2 facility). The average volume reduction factors generally range between 4 and 5.

COGEMA also operates a 2000 t supercompactor of a "more standard" design to treat low irradiating  $\beta\gamma$  waste.

Similarly, EDF, which formerly carried out low power compaction of its solid waste, operates an identical 2000 t supercompactor at its BUGEY plant to compact all the wastes from its nuclear power plants. The compacted drums are concreted in 400 l metal drums before shipment and disposal. This operation helps to condition 3 primary drums of 200 l per 400 l drum, at a rate of over fifteen 200 l drums compacted per hour.

### Incineration

This technology is very attractive considering the very high volume reduction factors available. Incineration technologies industrially developed by SGN are presented in a separate paper but we can stress that these incinerators are adapted both for incinerating solid wastes as well as liquid organic waste, thanks to the use of specific burners.

Their industrial capacities can be as high as 100/200 kg/h of waste and nuclearization operations have helped to adapt the SGN technology to the incineration of 20 kg/h of high-alpha waste.

Simultaneous development work is underway to optimize the conditioning of the ash produced, which is very often immobilized in matrices such as hydraulic binder or bitumen. To do this, SGN and the CEA have conducted joint R&D to design and industrialize ash fusion processes, particularly microwave processes which appear to be highly promising.

Research guidelines can also be observed in the area of solid waste incineration by offering additional volume reduction of the residue produced.

### Melting of metallic waste

To make further progresses in solid waste volume reduction, facilities were developed and constructed to process metallic waste as justified for instance by the 500 t of more or less contaminated steels arising only from the routine maintenance of the French nuclear power plants. Future decommissioning work will produce significant additional tonnages.

Two basic principles initially applied in conventional foundry are being implemented at a commercial scale : induction melting in a tilting crucible installed by CEA in a commercial prototype on the SACLAY site (500 kg metal/batch) and melting in a 3 electrode arc furnace installed at MARCOULE.

These processes are attractive since they allow maximum waste consolidation and produce ingots that could be reused in the nuclear industry, for instance for shielding. Evidence was also provided that melting contributes to decontamination work only by evenly distributing the activity initially present on the waste surface within the melted mass but also by separating certain radionuclides in the slag floating on the surface.

#### 4. Solid Waste Packaging

Immobilization of the processed solid waste is an integral part of their management in order to contain radioactive materials in a waste form which can maintain its integrity over very long period of time, thus effectively isolating the materials from the environment and hence from the public.

SGN acquired the full mastery of immobilization grout preparation, injection, and containers filling level control by numerous industrial applications, the most recent being operated at the La Hague reprocessing plant for hulls and caps (T1 facility) and active solid waste (AD2 facility) embedding into concrete.

As far the container itself is concerned, a multiple year research effort by COGEMA culminated in the development of a new process to immobilize nuclear waste in concrete containers reinforced with metal fibers.

The fiber reinforced concrete containers satisfy all French safety requirements relating to waste immobilization and disposal and have been certified as "high performance containers" by ANDRA, the national radwaste management agency.

COGEMA chose the "Fibraflex" fibre developed and fabricated by a company of the Saint-Gobain group. The patented manufacturing process gives Fibraflex its flexibility and strong mechanical properties while making it highly workable and its corrosion-resistant properties tested in comparison to be conventional carbon steel rebars.

Based on the results of the characterization programs, two types of packages received ANDRA certification :

- a cylindrical one, and
- a cubical one whose overall dimensions are:  $1.7 \times 1.7 \times 1.7\text{m}$ .

These containers have been fabricated on a production scale since July 1990 by SOGEFIBRE, a jointly-owned subsidiary of SGN and Compagnie Générale des Eaux in a specific workshop producing 12,000 containers per year.

SGN and SOGEFIBRE are now exporting fiber reinforced concrete technology presently in the USA and in Slovakia and are preparing to market fiber reinforced products to other clients for both nuclear and non-nuclear applications.

# **AN EXAMPLE OF A COMPLETE TREATMENT CYCLE FOR LOW- AND MEDIUM-LEVEL WASTE AT THE NUCLEAR RESEARCH CENTRE AT PUPSPITEK-SERPONG (INDONESIA)**

M. J. EYMERI    J. C. GAUTHEY  
TECHNICATOME, FRANCE

## **1. Introduction**

Nuclear power installations and nuclear research laboratories produce large quantities of liquid and solid waste.

So as to reduce the amount of waste to be stored and ensure that this is stored and warehoused in satisfactory environmental security and safety conditions, selective treatment, adapted to the type of waste, is performed in specialized installations.

For several years now TECHNICATOME has been developing, building, and putting into operation treatment and packaging processes adapted to each type of waste. By controlling the entire waste treatment process from production to storage, TECHNICATOME has been able to set up for BATAN (BADAN TENEGA ATOM NASIONAL) the Indonesian nuclear research centre, a radioactive waste treatment plant on the PUPSPITEK site.

The installation was built in collaboration with BATAN, thus ensuring a transfer of the technologies required to enable the Indonesian staff to operate the installation.

## **2. Presentation of the Nuclear Research Center**

Within the PUPSPITEK scientific center (research, science, and technology center) BATAN (The Nuclear Energy Commission) is responsible for studying, developing, and implementing nuclear energy processes. For this purpose, it is equipped with:

- a multi-activity reactor,
- a radioisotope production laboratory, and
- a reactor fuel production laboratory.

Waste produced by these different installations is treated by a waste treatment plant designed by TECHNICATOME and built in collaboration with Indonesian companies.

## **3. Organization of the Overall Project**

This was performed as part of a joint project between TECHNICATOME and BATAN:

- TECHNICATOME was responsible for overall engineering, supplying specific nuclear equipment, monitoring assembly, and tests, and
- BATAN was responsible for infrastructure and erection and putting the equipment into operation.

## **4. Installation Design Principles**

### **4. 1 Waste characteristics**

Waste produced by the research installation located on the Centre comprises:

- liquid waste with an activity of under  $5.4 \times 10^8$  Bq/m<sup>3</sup> ( $2 \times 10^{-2}$  Ci/m<sup>3</sup>),
- semi-liquid waste (ion exchange resins : IER) with a maximum activity of  $3.7 \times 10^9$  Bq/m<sup>3</sup> ( $0.1$  Ci/m<sup>3</sup>),
- solid technological waste (plastic bags, gloves, cotton, paper) packaged in 100 l metal drums.  
This waste has been sorted by the producer:
  - by type of waste (compactable, non-compactable, alpha emitter), and
  - by activity, so that, on contact, the dose rates of the packaged drums do not exceed 0.25 m Gy/h (25 mRad/h),
- biological waste with a maximum activity of  $3.7 \times 10^5$  Bq/kg ( $10^{-5}$  Ci/kg) packaged in plastic bags, and
- items of clothing (clothing, masks) packaged in 5 to 10 kg plastic bags with a dose flowrate of under  $2.5 \times 10^{-2}$  mGy/h (2.5 mRad/h).

### **4. 2 Installation functions**

The main purpose of the installation is to:

- treat radioactive waste with a view to producing a waste package which complies with prevailing storage standards; and
- carry out research and development work in the field of waste treatment.

#### **4. 2. 1 Waste treatment**

To ensure that waste treatment is carried out, all the equipment should be able to ensure the following functions:

- collect waste from the different producers and transport it to the treatment station;
- treat liquid waste by:
  - evaporation (liquid waste), or incineration (combustible waste), and
  - packaging in a concrete matrix;
- treat solid waste by:
  - compacting (compactible waste), incineration (combustible waste) or placing in a concrete shell (other solid waste),
  - packaging in a concrete matrix;
- treat semi-liquid waste by:
  - chemical pre-treatment,
  - packaging in a concrete matrix, and
  - treating items of contaminated clothing in a nuclear laundry;
- store the packages produced before they are transported to the repository; and
- treat clothing.

#### **4. 2. 2 R&D studies**

R&D studies are performed in the laboratories.

### **4. 3 Basic Design Principles**

The installation comprises three buildings:

- the main building which houses the process and administration areas,
- a building which groups together the auxiliary units (various utilities, generator), and
- a building which includes the package storage area.

The main building comprises two blocks (see Appendix 1):

- one "cold" block which groups together the administration building, general technical areas, and the centralized control room,
- one "hot" block sub-divided into distinct areas corresponding to each type of process; each area has its own control room.

The entrances and exits to/from the "hot" block are through distinct air locks:

- "hot" and "cold" staff cloakrooms, and
- an equipment hatch specific to each area.

## 5. Description of the Installation

### 5. 1 General information

The circuit, followed by the waste immediately arriving at the station, depends on the type of waste. The table provided in Appendix 2 shows the succession of operations carried out for each type of waste.

### 5. 2 Treatment of aqueous liquid waste

#### 5. 2. 1 Purpose of treatment

Treatment by evaporation makes it possible to obtain:

- a concentrate which contains all the active matter and is immobilized in a concrete phase, and
- a distillate which is free from active matter and discharged into the environment.

#### 5. 2. 2 Transport to the plant

Liquid waste is transported from the storage tank (reactor, laboratories, workshops) from the producers in the plant to the installation in a 8 m<sup>3</sup> tanker (cf Appendix 3).

Tanker design complies with the regulations governing the transport of dangerous materials in France. The vehicle is therefore authorized to use the public highway.

It is fitted with an autonomous pumping system which ensures:

- either the filling of the tanker using the producer tanks,
- or drainage into the treatment plant tanks.

A control-command desk located in the lateral section enables the operator to pilot transfer operations from the truck.

The tanker is fitted with all biological protection and safety devices (retention troughs, gas filtration, etc.), ensuring safety of operations and protection for the operating staff.

#### 5. 2. 3 Waste storage

This comprises four 50 m<sup>3</sup> tanks each equipped with its own homogenization system:

- two tanks are used to receive waste from the tanker. The choice of whether one or the other tank should be filled depends upon the type of waste;
- one tank is used to supply the evaporator;

- one tank, which normally remains empty, is used as standby:
  - either to collect waste with specific characteristics,
  - or to perform mixing operations to ensure that the physicochemical characteristics of the waste comply with those required for the evaporator.

Tank functions are interchangeable.

A pumping system is used for inter-tank transfers and to supply the evaporator.

The tanks are located in a specific individual room connected to nuclear ventilation.

#### **5. 2. 4 Evaporation**

##### **5. 2. 4. 1 Description of the installation**

Installation capacity is 750 l/h. The volume reduction ratio is 50.

Dry extract concentrations in the condensate and distillate are 250 g/l and  $2.5 \times 10^{-4}$  g/l respectively.

It comprises:

- a tubular exchanger with thermosiphon circulation supplied with waste to be evaporated. It is heated using live steam to ensure the vaporization of liquid waste;
- a tranquilization tank at the exchanger outlet which is used to separate the liquid and steam phases. The steam phase is directed to the separation column. The concentrate obtained is removed from the base of the tank;
- a separation column. The liquid carried into the steam phase is separated from this steam by washing through the exchanger plates using a back flow of demineralized water; and
- a condenser which ensures condensation of the steam phase and a cooler which is used to cool the distillates.

The evaporator is fitted with an anti-foam in-service injection system to prevent the formation of foam which could impede the separation of the steam and liquid phases.

##### **5. 2. 4. 2 Storage of distillates and concentrates**

The distillates are stored in two 50 m<sup>3</sup> tanks. They are fitted with sampling and chemical, and radiochemical measuring systems.

In the event of an evaporator malfunctioning, an automatic system sends the distillate into the front tanks.

A 3 m<sup>3</sup> tank fitted with a sampling system recovers the concentrates before they are transferred to the conditioning unit.

##### **5. 2. 4. 3 Control principles**

With the exception of the chemical reagent circuits, which are controlled locally, the system is controlled from the evaporator control room where the staff has at its disposal:

- a mimic diagram which groups together all the signals,
- the indicators and recorders required to monitor the system,
- control buttons for the actuators and remote controlled valves, and
- the transmission of alarms indicating threshold overshoots or operational defects.

Evaporation operations are automatic. In the event of a malfunctioning, the safety devices set the installation in standby.

### **5. 2. 5 Waste packaging**

#### **5. 2. 5. 1 Description of the installation**

This cubicle is equipped to package the following wastes into concrete shells :

- evaporation concentrates,
- ion exchange resins, and
- solid non-compactable waste and 200 l drums of already compacted waste.

They undergo the following operations :

- controls using sampling and laboratory analysis to check the physicochemical and radiological characteristics of the waste,
- chemical pretreatment, if required (e. g. concentrates and ion exchange resins),
- transfer and dosing using the capacities specific to each waste (e. g. concentrates and IER), and
- final mixing in the shell (IER and concentrates) or blocking by injecting a concrete grout (non-compactable or compacted waste).

The installation comprises additional circuits for storing, weighing, and transferring the dry concrete grout components and a concrete grout preparation unit.

The concrete shells are removed to a room where :

- the concrete is set, and
- a protection plug is made.

When these operations are completed, the shells are transferred to the storage room after radiological control.

### **5. 2. 5. 2 Control principles**

The unit is designed to operate 8 hours per day. Two concrete shells are produced each day.

Operations are controlled from a control desk located in the control room where the staff has at its disposal :

- a mimic diagram which groups together all the signals, and
- control buttons for the actuators and remote controlled valves.

Concreting operations are automatic. In the event of a malfunctioning, the safety devices sets the installation in standby.

### **5. 2. 6 Storage**

In the storage room :

- the concreted shells are stored,
- the shells and drums of waste from other waste production centers are unloaded, and
- the shells and drums are loaded to be transported to the final storage centre.

It is divided into three main areas :

- shell storage,
- drum storage, and
- drum and shell loading and unloading.

Storage capacity is 1500 drums and 500 concrete shells. These are handled using a crane and pallet stackers.

### **5. 3 Treatment of solid waste**

#### **5. 3. 1. Purpose of treatment**

The purpose of solid waste treatment is to treat:

- non-combustible waste by compaction, and
- combustible waste by incineration.

The waste which is obtained (incineration ash, compaction slabs) are then packaged into drums or concrete shells in a concrete matrix.

#### **5. 3. 2 Transport to the installation**

The waste is transported by truck from its production site to the installation. It is packaged in 100 l metal drums. Waste which can be incinerated is separated, on the production site, from waste which cannot.

#### **5. 3. 3 Storage before treatment**

In a specific room storage is provided for:

- the 100 litre drums of non-combustible waste,
- the empty 200 l drums into which the first drums are compacted, and
- the 100 litre drums of combustible waste.

#### **5. 3. 4 Compaction of drums containing non-combustible waste**

The 100 l drums of solid waste which cannot be incinerated are compacted into the 200 litre drums using a 600 kN hydraulic press. The installation is designed to produce 5200-litre drums per day.

The press is fitted with a nuclear ventilation circuit ensuring that containment is maintained during operations.

The installation is controlled and operated . . . manually and locally.

#### **5. 3. 5 Packaging the compacted units**

After the 200 litre drums are filled, they are then filled with cement grout and vibrated to ensure that the compacted waste is packaged (see Paragraph 5. 2. 5).

#### **5. 3. 6 Incineration of combustible waste**

##### **5. 3. 6. 1 Description of the installation**

The incineration unit ensures the incineration of:

- Solid combustible waste:

- cotton,
- polyethylene, and
- PVC;

- burnable liquid waste:

- oil,
- scintillation liquids, and

- tributyl phosphate (TBP) diluted in dodecane; and
- biological waste.

Installation capacity is 50 kg/h for solid waste and 20 kg/h for liquid waste.

The 100 l drums of combustible waste are transferred from the 100 and 200 l drum storage room to the incineration room (cf Paragraph 5. 3. 3).

The contents of the drums are packaged into cardboard boxes containing 3. 7 kg. This is done in glove boxes.

The cardboard boxes are loaded automatically into the incinerator through a double air chamber.

The liquid waste to be incinerated is stored in a 1 m<sup>3</sup> tank (waste not containing TBP) and in a 0. 1 m<sup>3</sup> tank (waste containing TBP). It is transferred into the incinerator after possible dilution, so as to limit the quantity of TBP in the liquid waste to be incinerated.

The technological solid waste is packaged in plastic bags and stored in deep freezers..

It is then packed into cardboard boxes to be fed into the incinerator , in the glove boxes.

The installation comprises (see Appendix 4):

- a furnace composed of:

- a first chamber where oxygen reduced combustion is performed for solid waste (temperature between 800 °C and 1,000 °C), and
- a second chamber where post-combustion is performed using gas produced in the 1st chamber during solid waste and liquid waste combustion operations. The temperature in this chamber is approximately 1,000 °C to 1,200 °C,
- a dilution chamber to reduce the gas temperature to 180 °C by dilution in the ambient air,
- a two-stage gas filtration assembly:
  - approximate filtration in a sack filter with automatic unclogging using a continuous flow of compressed air, and
  - filtration of the radioactive particulates using the HEPA filters,
- a gas washing unit so as to:
  - cool the gas produced by filtration, and
  - neutralize the acid gases produced during combustion.

This unit comprises a venturi and a separation column.

- an extraction assembly comprising:

- a gas reheater, and
- a gas extraction ventilator.

Gas activity is continually monitored before it is discharged into the atmosphere.

### **5. 3. 6. 2 Control principles**

Waste incineration is performed in 6 day cycles. Each day includes a 6 hour period during which the unit incinerates waste and an 18 hour period during which the unit is maintained at a constant temperature (thermal standby).

Ash is removed from the furnace each day before the incineration period.

The unit is controlled from a control room separated from the incinerator room and equipped with a control panel with mimic diagram, indicators, and recorders.

The installation operates automatically. In the event of a malfunctioning, the safety devices set the installation in standby.

### **5. 3. 7 Packaging the incineration ash**

Ash is removed from the first combustion chamber and the bag filters through an air chamber, where it is cooled before being recovered, with continuous confinement, in a metal drum which has been previously filled with encapsulation materials.

The ash is immobilised in the cement using a drum-rotation mixer.

The drums are taken to a room (see Paragraph 5. 2. 5. 1) while the concrete sets, before being taken to the storage room after the packages have been radiologically controlled.

### **5. 4 Treatment of burnable liquid waste**

This waste is delivered in demi-johns and transferred to the burnable liquid waste storage area (see Paragraph 5. 3. 6. 1). The tanks containing liquid waste with TBP are differentiated from those without TBP.

The waste is then incinerated as described in Paragraph 5. 3. 6. 1.

### **5. 5 Treatment of semi-liquid waste**

#### **5. 5. 1 Purpose of the treatment**

The purpose of treating semi-liquid waste (ion exchange resins) is to immobilise it in a concrete matrix.

#### **5. 5. 2 Transport to the installation**

The ion exchange resins are carried from the storage tanks of the center producers to the installation in a 1. 5 m<sup>3</sup> tanker. Its design criteria are the same as the ones for the liquid effluents (see Paragraph 5. 2. 2).

#### **5. 5. 3 Storage of the ion exchange resins**

The ion exchange resins are transferred by a hydraulic system into a 5 m<sup>3</sup> tank where they are stored before packaging.

#### **5. 5. 4 Packaging ion exchange resins and removing the packages**

This is described in Paragraphs 5. 2. 5 and 5. 2. 6.

### **5. 6 Treatment of laundry**

#### **5. 6. 1 Aim of the treatment**

The laundry to be treated comprises working clothes and gas masks used on the site installations.

This treatment is aimed at:

- sorting the contaminated laundry for treatment as solid waste (incineration or compaction), and
- cleaning of non-contaminated laundry, for re-use by the staff.

#### **5. 6. 2 Description of the installation**

Laundry is sent to the station packaged in plastic bags in 100 l drums containing a 5 to 10 kg

load per drum.

Preliminary sorting operations are performed by measuring the dose rate on contact with the plastic bags, thus making it possible to direct bags with a dose rate greater than the specified values to the solid waste treatment system.

The unit is composed of the following equipment:

- two washing machines,
- two rotary driers,
- two ironing presses fitted with steam heating systems, and
- two glove boxes used when opening the plastic bags.

After decontamination in the washing machines, the laundry is dried in the rotary driers.

Water produced during decontamination is transferred to the waste storage tanks.

Radiological controls are used to separate any non-contaminated clothing treated as solid waste.

Decontaminated laundry is ironed using the ironing presses. A sewing machine is used for everyday mending operations.

The washing machines and glove boxes are maintained in dynamic confinement.

#### **5. 6. 3 Control principles**

The washing machines are controlled locally by an operator. Washing machines and driers operate automatically. The sorting machine operates semi-automatically (continuous manual loading, measuring, and automatic sorting).

#### **5. 6. 4 Removing the clean laundry**

Once decontamination is completed, laundry is re-allocated to the different units.

#### **5. 7 R&D Laboratories**

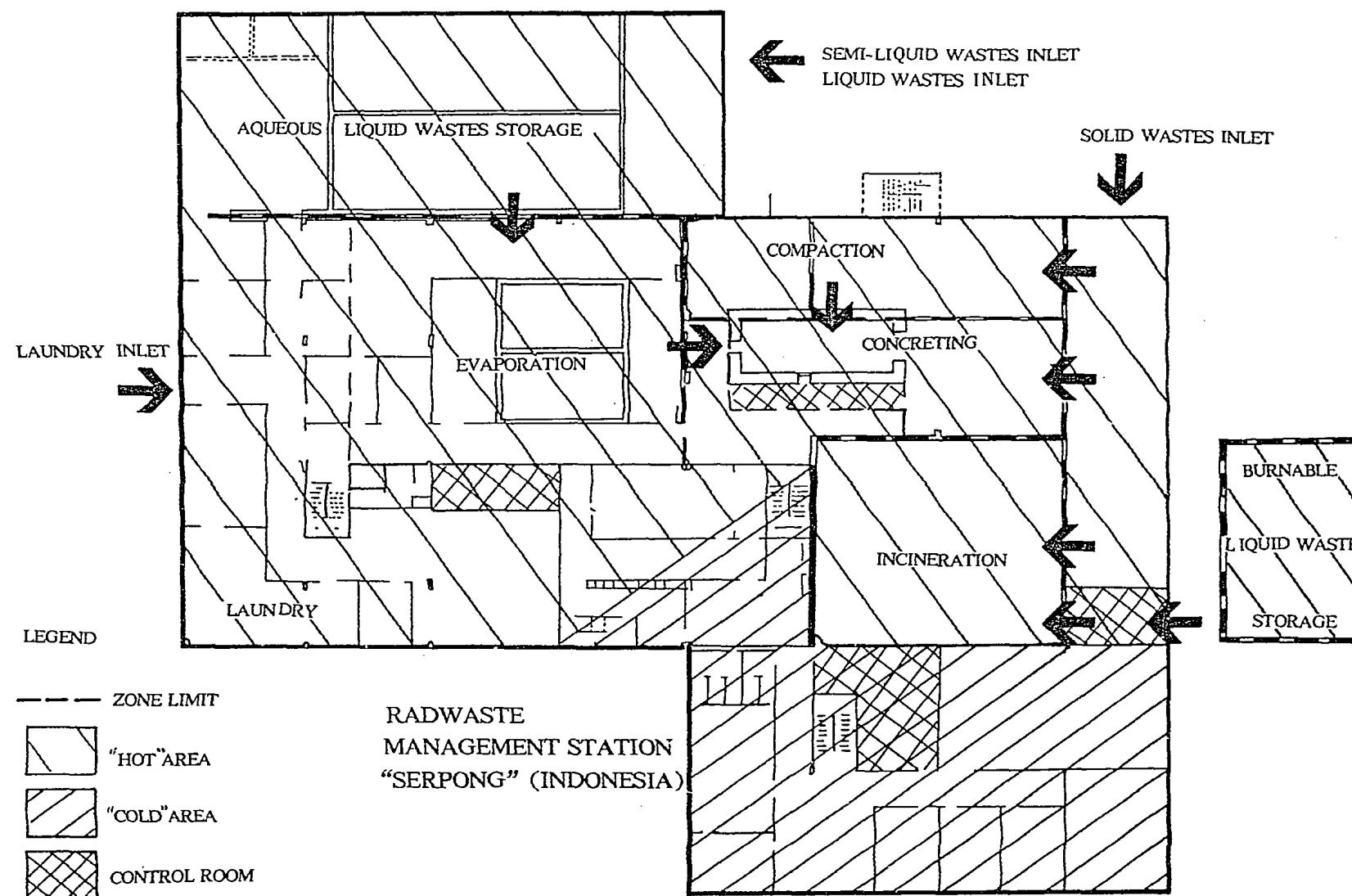
The aim of these laboratories is:

- to run R & D waste treatment operation, and
- to run routine physicochemical evaluation operations.

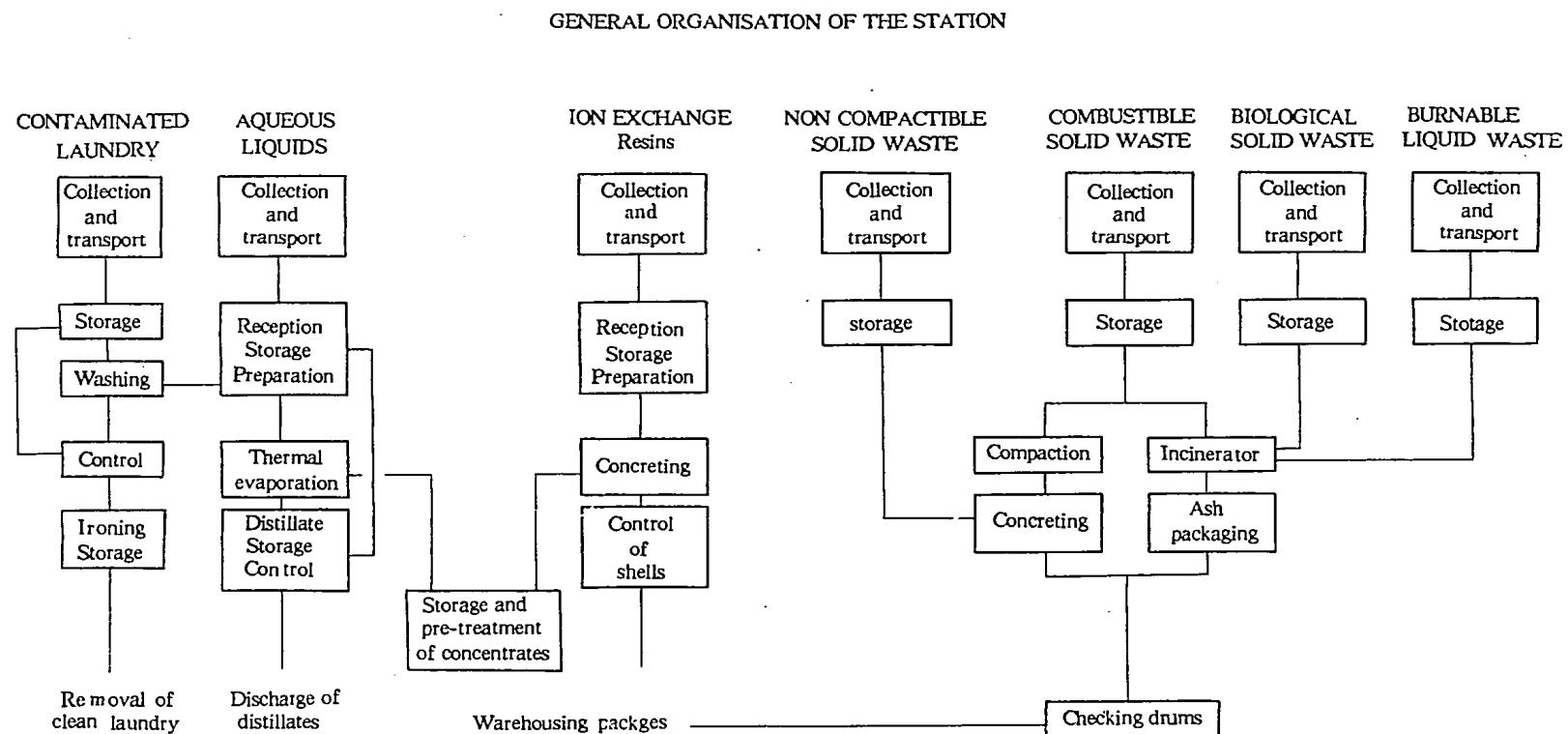
They are equipped with standard physics-chemistry equipment:

pH-meters, spectrophotometers, conductivity meters, spectrometers, etc.

Appendix 1 Definition of Installation Zones

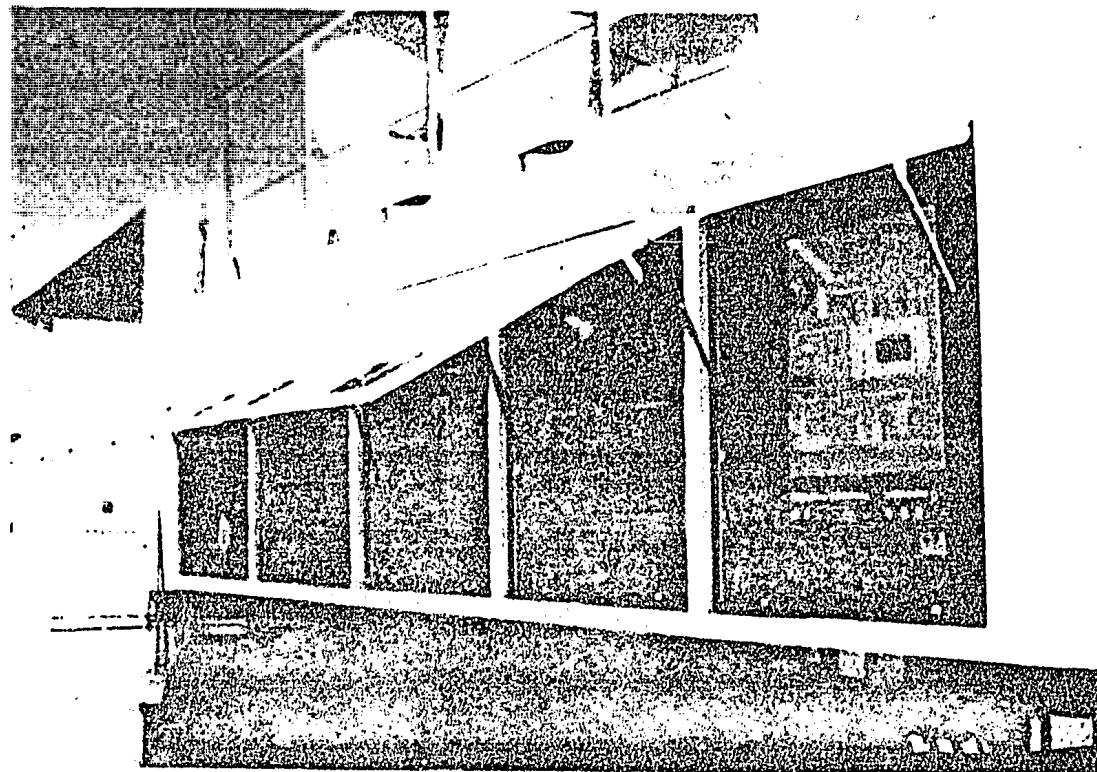


## Appendix 2 Functional Description of the Installation

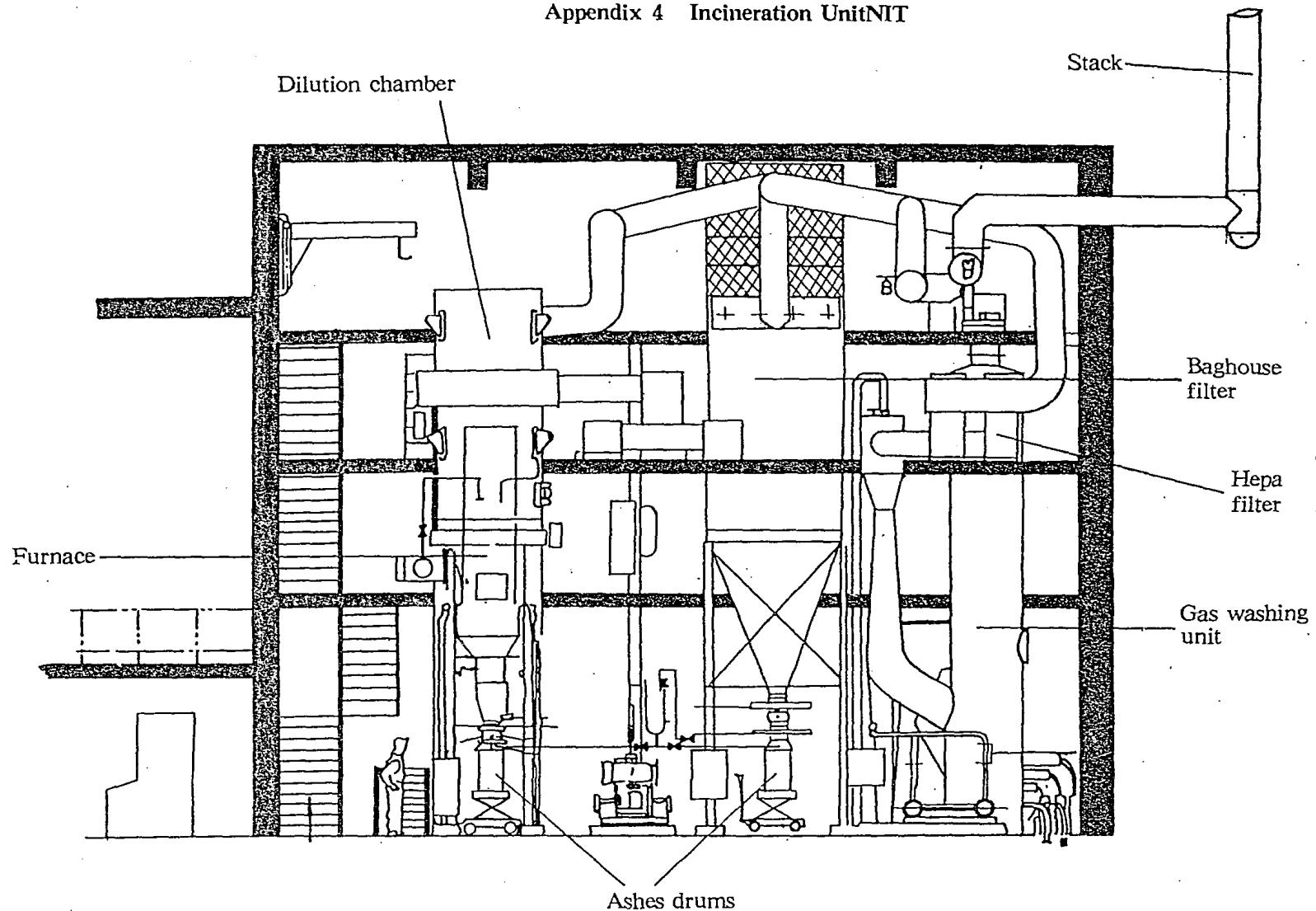


**Appendix 3 Tankers Trailors Liquid Waste**

**TANKERS TRAILORS  
LIQUID WASTE**



## Appendix 4 Incineration Unit NIT



# **CHARACTERIZATION OF LOW AND MEDIUM ACTIVE WASTES**

**ARSENE SAAS  
CEA, FRANCE**

## **Introduction**

The characterization of a material or an object as waste, a matrix, a coating or a package, consists of determining its specific properties.

In the characterization of nuclear waste, we restrict ourselves to the acquisition of characteristics which have to be known in order to evaluate safety, and temporary and permanent storage conditions.

### **1. Objectives of Waste Characterization and Characterization Types**

Five main objectives are looked for when performing a characterization test. The tests are intended to :

- guarantee the mechanical properties of the coated waste and the matrix,
- test the durability of the physical and mechanical properties of the waste for the storage duration,
- evaluate the behavior of isotopes in the waste and the long-term waste-matrix interaction,
- calculate the loss rate by isotope leaching, and
- compare the results with specifications, and rules specified by their Authorities and Safety notices.

For FRANCE, this involves ANDRA specifications, the Basic Safety Rules (*les Règles Fondamentales de Sûreté—RFS*) of the Central Service (Ministry of Industry), Safety Instructions from the Nuclear Safety and Protection Institute (*l'Institut de Protection et de sûreté Nucléaire—IPSN*).

Several types of characterization are necessary to cover all knowledge useful for storage and repository of radioactive waste.

There are three main types of characterization :

- characterization of packages and materials,
- behavioral studies, and
- long-term behavioral studies.

#### **1. 1 Characterization of packages and materials**

This type of characterization can be defined as the acquisition of the characteristics of a coating, a package or a material in its initial state; it is therefore the creation of a statement. This type of characterisation is carried out in three steps:

- characterization of the waste itself,
- pre-characterization of the material and the coating, and
- characterization of the finished product (the package intended for storage or repository).

### **1.1.1 Characterization of the waste**

This is defined as acquisition of data necessary for :

- producing the waste package,
- classification for its activity and its destination:
  - Class FA-MA-HA ,
  - Class  $\beta$ ,  $\gamma$  or  $\alpha$ , and
  - destination: surface repository, temporary storage before repository at depth; and
- analysis of basic safety and its entry into the approval file.

### **1.1.2 Pre-characterization (process and coating)**

This is the essential stage for the evaluation of the quality of a process and the coating resulting from all the pre-production steps (formulation, reference tests, terminal tests, sensitivity test, etc.).

Tests for this stage are carried out using reference materials.

Two types of objective are involved :

- a comparison of the various performances of the coating (choice of matrices, coating rate, formulation, etc.), and
- a demonstration that the coatings, the packaging, and the finished products satisfy standards and specifications in force.

This step normally results in the production of a packaging specification and the production of nominal specifications for coated waste.

### **1.1.3 Characterization of the finished product**

This stage involves three types of investigation :

- examination of the intrinsic characteristics of packagings and coatings,
- checking the durability of these characteristics (degradability, confinement, etc.), and
- defining the influence of variations of the various process parameters (sensitivity tests) in order to guarantee the quality of the coated material.

Characteristics and acceptance criteria for the finished product are examined within Chapter 2 of this document.

## **1.2 Behavioral studies**

This second section of the characterization of nuclear waste concerns the *source term* study. It is defined as being the acquisition of basic data for forecasting and modelling isotope emission in storage environments.

Two final results are sought in this type of characterization :

- understanding the phenomena involved in isotope emission, and
- the qualitative and quantitative expression of the basic values and ranges of values necessary for the storage safety analyses.

## **1.3 Long-term behavior**

This type of characterization is defined as the acquisition of data for mechanisms dealing with *slowly evolving* phenomena. Two approaches can be used :

- the behavior of natural counterparts, and

- the thermodynamic definition of slow phenomena.

The final result that is expected to be obtained from this last characterization stage consists of changing from "high bound" type modelling to a modelling system based on extrapolating variations of characteristic, making use of relevant measurements and bounding values.

## 2. Package Approval: Specifications, Procedures, and Acceptability Criteria

Inspection of the current situation in countries developing nuclear energy shows that most countries base approval of waste packages on three main data items:

- specifications produced by the authorities,
- procedures produced by approved laboratories and organizations, and
- acceptability criteria or quality criteria which should lead to the respect of specifications.

Although the organization of waste-related responsibilities and therefore the specifications issued by the various authorities vary between countries, all have established some characterization procedures and acceptability criteria for waste packages.

### 2.1 Procedures — Technical sheets

In France, approval of a waste package is announced after inspection of experimental results and tests in accordance with the minimum characterization program defined by ANDRA.

This minimum program affects the three selected activity levels and the two classes of waste packages (homogeneous-heterogeneous). Tests and checks are classified into four categories:

- physical characteristics,
- mechanical characteristics,
- confinement power, and
- stability-degradability (maintenance of confinement power).

In order to reply uniformly to the creation of tests required for the approval file, the Confinement Checking and Evaluation Office (Bureau d'Evaluation et de Contrôle des Confinements—BECC) has published a set of technical forms condifying a general method of executing these tests.

A list of currently available technical forms is given in Table 1.

#### SHEETS UNDER PREPARATION:

- Behavior on immersion in water,
- Exudation of water under compression,
- Homogeneity test of a coating,
- Non-destructive measurement of  $\gamma$  producers in real waste package, and
- Evaluation of the resistance of bitumen coatings to microorganism attack.

A block diagram has been produced by the Waste and Effluents Management for the approval procedure (Direction des Effluents et Déchets — Dg/ED). Figure 1 summarizes the global routing of an approval circuit and specifies the role of each participant for wastes which can be delivered to ANDRA on a surface site.

ANDRA approval of waste packages requires the production of the following documents:

- Description of the production process for the package containing the waste,

- Quality assurance program for the implementation of the package production processes as described in the process description, and
- Package characterization test report (technical characterization file).

The characterization test report assumes that the producer has previously submitted a draft characterization test program to ANDRA. When this draft has been accepted by ANDRA, it then becomes the *characterization procedure*, containing a description of the checks and tests to be carried out.

Therefore, the creation of a test requires the following definitions (see Figure 2):

- the sample or samples,
- the size and characteristics,
- the test procedure,
- the results to be provided, and
- the test report.

All reports for all tests described in the protocol must then be inspected and used to produce a *test summary report*, or a *characterization technical file* which is forwarded, as already stated, to ANDRA by the producer.

To illustrate the importance of this procedure, we will partially describe the procedure for measuring the initial activity of raw waste and coatings which meets ANDRA's requirement for the monitoring of the activity stored on the site in order to respect site storage capacity for each isotope.

Figure 3 shows ANDRA's mandatory considerations towards the safety authorities, and also the consequences of respecting mandatory quality requirements demanded by ANDRA for producers' packages.

Consequently, waste activity measurement (raw or coating) is one of the most important criteria. Characterization therefore requires the implementation of an adapted measurement scheme for each waste or coating type.

Each type of waste has its own time schedule for taking measurements and for monitoring the procedure so as to satisfy safety authority requirements.

The main procedure for coatings is shown in Figure 4. This Figure clearly shows the two recommended types of measurements:

- non-destructive measurements, and
- destructive measurements with the various related determinations.

Finally, to complete the execution of these measurements, Figure 5 defines the essential elements which must appear in the test report intended for the producer in order to prepare the summary report for all the tests, as required by the final management authority (ANDRA for FRANCE).

## 2.2 Package acceptability criteria

For FRANCE, characterization deals only with coating and package quality criteria. There are listed in the *minimum characterization program* defined by ANDRA as recalled in Section 2.1.

In view of the available characterization results for various matrices enclosing various types of waste, the BECC established a summary of coating and packaging qualities, as a function of the

main parameters.

Three types of parameters were used for this summary (Table 2) :

- quality of the coating-quality of package (RFS+ANDRA specifications),
- implementation and feasibility , and
- control.

An evaluation was made for each type of parameter, matrix, and waste type (homogeneous-heterogeneous) based on the acquired characterization results.

## Conclusions

For several years now, research on raw or packaged waste characterization has been carried out in France, in particular those aspects concerning :

- legislation ,
- regulations,
- standardization of methods and techniques, and
- laboratory and test hall equipment.

The BECC provides the main communication link between the waste producer, ANDRA, and the CEA which can perform all services related to tests and corresponding measurements, either as part of its own research and development program, or on request by the BECC. The CEA resources, methods, laboratory, and test halls, which are available to the BECC and its customers, now form a characterization system adapted to each waste or packaging type to which a quality assurance program is applied.

BECC manufacturing experience enables it to give correct advice to the waste manufacturer and any nuclear material safety unit, both on the selection of packaging and test procedures, and on equipment and the selection of characterization materials or the organization to be set up. In particular, the BECC can provide the following services:

- qualitative or quantitative analysis of radionuclides present in already packaged waste (including badly packaged waste), or unpackaged waste; and, as far as possible, evaluation of the main physico-mechanical and confinement characteristics,
- technical assistance for the characterization of packages including:
  - developing specific technical test forms according to the customer,
  - establishing BECC type characterization coordination units and training corresponding personnel,
  - transferring basic elements for the implementation of a quality assurance program for package characterization, and
  - establishing basic characterization method and packaging expertise planning, and
- technical assistance with the design and construction of characterization laboratories.

The Figure 6 shows a summary of the main missions provided by the BECC from its experience.

To conclude, we would like to emphasize that the objective of the various units in charge of characterization is to supply technical assistance in view of the harmonious development of waste approval files and the development of test techniques to improve knowledge of the intrinsic characteristics of packaged waste. Above all, the framework in which these characterization activities are carried out must respect the special features of this characterization, which is at the

very interface of the various radioactive waste management partners: safety authorities, waste management (ANDRA), waste producers, and the managers of research and development programs.

Table 1 Technical sheets for FA-MA tests

01. PHYSICAL CHARACTERISTICS

<u>Form number</u>	<u>Title</u>
FT 01. 001	Measurement of the apparent density
FT 01. 002	Determination of the flash point of polymer-based coatings
FT 01. 003	Determination of open vessel fire points on bitumen-based coatings
FT 01. 004	Determination of the spontaneous ignition point of bitumen coatings
FT 01. 005	Exothermicity test on REI coating by a thermosetting resin

02. MECHANICAL CHARACTERISTICS

<u>Form number</u>	<u>Title</u>
FT 02. 003	Determination of the crushing strength
FT 02. 004	Determination of the shock resistance
FT 02. 006	Determination of a coating's fire resistance
FT 02. 010	Determination of the compressive strength (hydraulic binder)
FT 02. 011	Determination of the compressive strength (polymer)
FT 02. 031	Determination of the resistance to thermal effects

03. CHEMICAL CHARACTERISTICS

<u>Form number</u>	<u>Title</u>
FT 03. 001	Determination of the water content of a bitumen coating
FT 03. 002	Determination of the content of salts or REI of bitumen coating

04. CHARACTERISTICS RELATED TO CONFINEMENT POWER

<u>Form number</u>	<u>Title</u>
FT 04. 001	Determination of the porosity of a coating to water
FT 04. 002	Measurement of the permeability to water of material based on hydraulic and/or polymer binders
FT 04. 003	Measurement of the permeation of a gas through a bitumen membrane or a bitumen coating
FT 04. 004	Determination of the porosity to mercury of coatings, coating materials, and waste
FT 04. 011	Measurement of the permeability to gas (nitrogen, helium, etc.) for hydraulic binders
FT 04. 020	Determination of leaching resistance
FT 04. 021	Measurement of radionuclide transfer measurement in hydraulic binder based materials. Application to the confinement of heterogeneous wastes

05. RADIOACTIVE CHARACTERISTICS

<u>Form number</u>	<u>Title</u>
FT 05. 030	Effects of $\beta$ 、 $\gamma$ radiation
06. GENERAL METHODS	
<u>Form number</u>	<u>Title</u>
FT 06. 001	Sample sizing
FT 06. 002	Expression and introduction of leaching test results
FT 06. 003	Determination of report on samples after leaching test
FT 06. 004	Ao polymers, raw or coated waste
FT 06. 005	Sample identification and monitoring for characterization

Table 2 List of parameters and criteria

1. QUALITY OF COATING - QUALITY OF PACKAGING (RFS-ANDRA SPECIFICATIONS)

1. 1 Physical characteristics

- porosity,
- permeability to water,
- permeability to gas,
- coating density,
- shrinkage,
- state change temperature,
- homogeneity of coated or blocked waste,
- homogeneity of coating,
- percentage of contained water,
- exudation of water in the waste under compression force,
- solubility in water of the coating-overcoating material, and
- filling rate.

1. 2 Mechanical characteristics

- compression strength,
- behavior of the packages under load, and
- package shock resistance.

1. 3 Confinement power

1. 4 Stability - Degradability - Maintenance of confinement power

- temperature cycle,
- behavior under radiation,
- sensitivity to contact with water,
- packaging resistance to corrosion in a humid environment, and
- package behavior under fire.

2. IMPLEMENTATION AND FEASIBILITY

2. 1 Industrial process

- Type of industrial installation: fixed or mobile installation,
- coating or blocking methods: continuous-discontinuous process, and

- installation capacity: high capacity  $> 50 \text{ m}^3/\text{year}$ , low capacity or pilot  $< 50 \text{ m}^3/\text{year}$ .

## 2.2 Process capacity for improvement

Related to the process itself: operating parameter,

- related to formulation: addition, and
- related to package produced: package dimension.

## 2.3 Additional packaging and safety

- Inflammability of basic materials,
- need for overcoating, and
- special packaging.

# 3. CONTROL

## 3.1 Procurement of basic materials

## 3.2 Storage conditions of basic materials

## 3.3 Cost price of basic materials

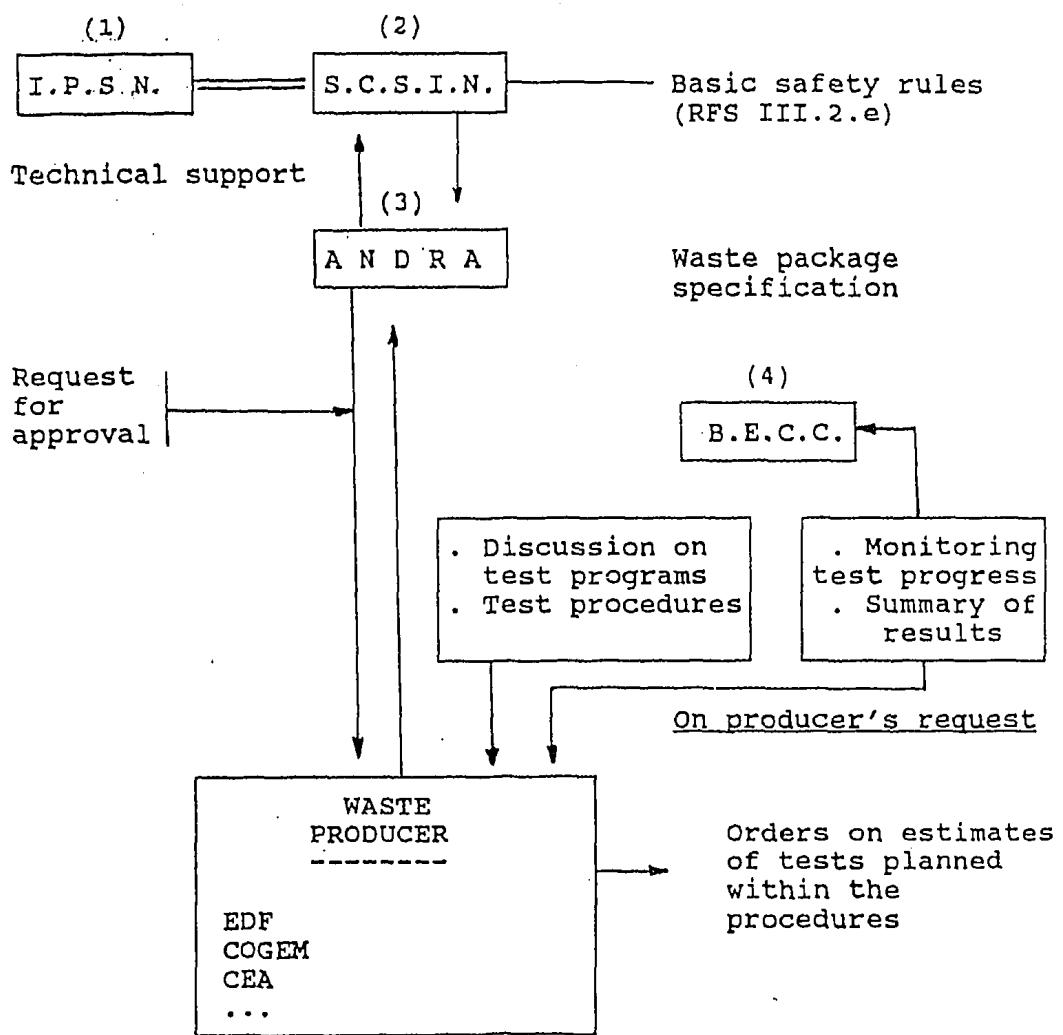
## 3.4 Constraints related to use of the process

- Energy cosumption ( $8^\circ$ ), and
- Constraints related to the number of parameters to be considered.

## 3.5 Storage - Handling

- Core-polymerization time before delivery, and
- Storage conditions on the facility.

Fig. 1 Characterization of Waste Packages  
for Delivery to ANDRA



(1) IPSN : Institut de Protection et de sûreté Nucléaire (CEA).

(2) SCSIN : Service Central de sûreté des Installations Nucléaires (Ministry of Industry).

(3) ANDRA : Agence nationale pour la Gestion des Déchets Radioactifs (National Agency for Radioactive Waste Management).

(4) BECC : Confinement checking and Evaluation Office (attached to the Service dealing with the characterization, confinement evaluation, and analysis at CEA).

**Fig. 2 The HE Characterization Procedure**

**CONTENTS**

- MAIN PROCESS DATA (COATING - WASTE COMPOSITION - GENERAL PERFORMANCES),

- DESCRIPTION OF CHARACTERIZATION TESTS:

- FOR THE COATING:

**INACTIVE TESTS**

LIST OF TESTS: NUMBER OF SAMPLES

DIMENSION

PREPARATION

TECHNICAL FORM REFERENCE

**ACTIVE TESTS (DITTO)**

\* FOR THE MATRIX AND THE WASTE (POSSIBLY)

\* FOR THE PACKAGE:

FILLING RATE

OTHER TESTS: FIRE - DROP - LEACHING - THERMAL CYCLE, etc.

- PRESENTATION OF RESULTS - TEST REPORTS

- EXECUTION SCHEDULE

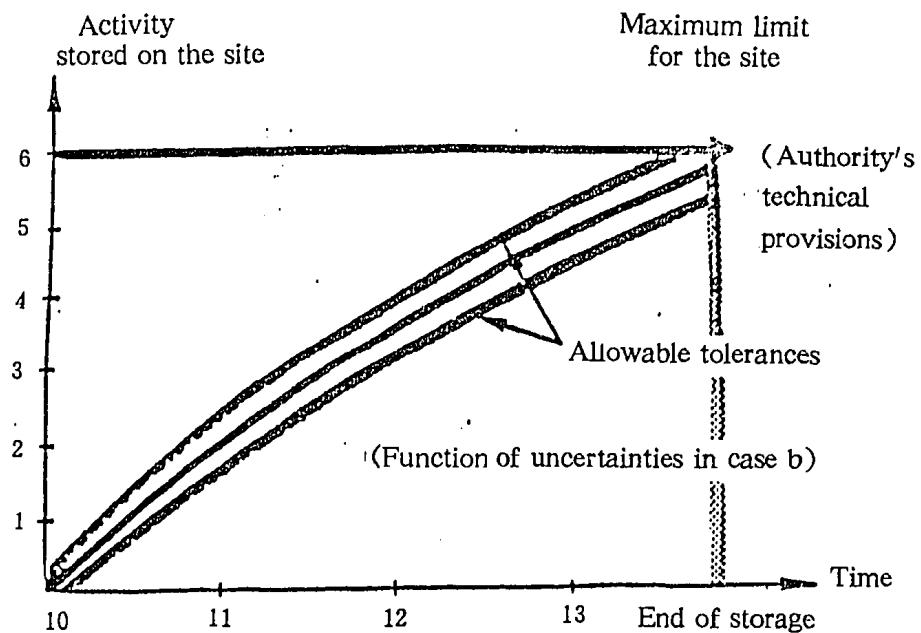
- REFERENCES: PROCESS BOOKS

ANDRA SPECIFICATIONS

RFS

Fig. 3 Producer's and ANDRA's Objectives

Case a) Manager-Authority



Case b) Producer

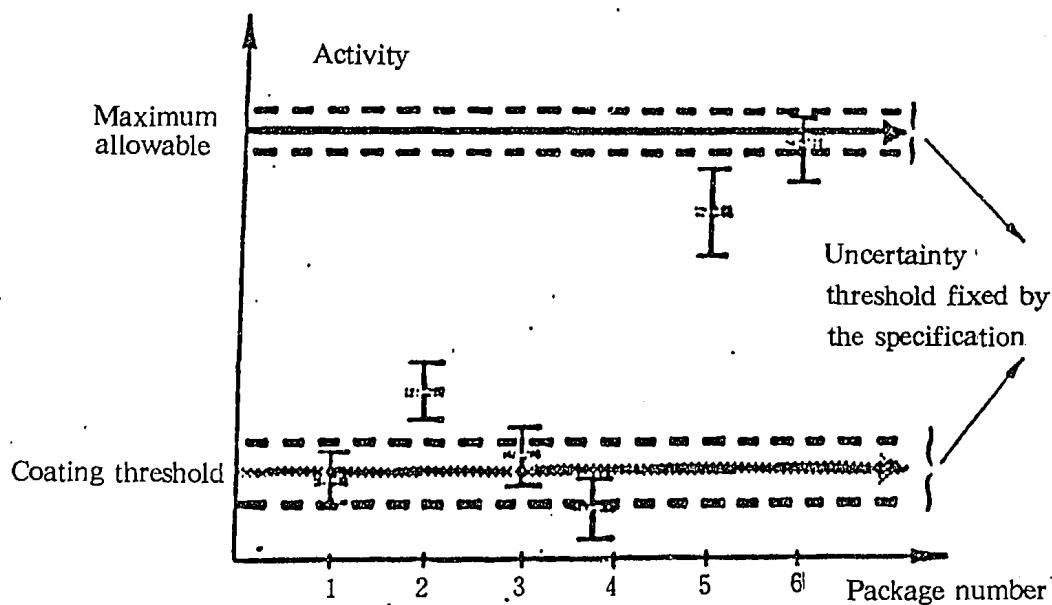
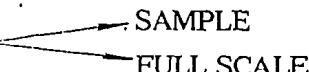


Fig. 4 Measurements of Coated Homogeneous Wastes

1. BITUMENS  
OUTLINE

1) NON-DESTRUCTIVE MEASUREMENTS 

2) DESTRUCTIVE MEASUREMENTS

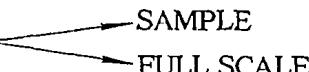
SALT EXTRACTION

BITUMEN PHASE ACTIVITY CHECK

$\beta\gamma$ ,  $\alpha$ ,  $\beta$  MEASUREMENTS ON SALTS

CHEMICAL MEASUREMENTS

2. POLYMERS  
OUTLINE

1) NON-DESTRUCTIVE MEASUREMENTS 

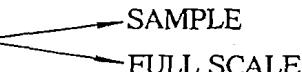
2) DESTRUCTIVE MEASUREMENTS

→ MINERALIZATION - DISSOLUTION ( $HNO_3$  -  $H_2O_2$ )

→  $\beta\gamma$ ,  $\alpha$ ,  $\beta$  MEASUREMENTS

→ SPECIAL CHEMICAL MEASUREMENTS (BORON, ETC.)

3. CEMENTS  
OUTLINE

1) NON-DESTRUCTIVE MEASUREMENTS 

2) DESTRUCTIVE MEASUREMENTS

MINERALIZATION - DISSOLUTION ( $HNO_3$  - HF)

$\beta\gamma$ ,  $\alpha$ ,  $\beta$  MEASUREMENTS

SPECIAL CHEMICAL MEASUREMENTS (BORON, Cl, ETC.)

### Fig. 5 Report

THE LABORATORY WILL ESTABLISH AND PROVIDE THE FOLLOWING MINIMUM INFORMATION :

- SAMPLE IDENTIFICATION,
- MEASUREMENT DATE,
- DATE OF THE MOST RECENT INSTRUMENT CALIBRATION,
- RESULTS WITH UNCERTAINTIES,
- FOR DESTRUCTIVE MEASUREMENTS, THE PRINCIPLE OF THE METHOD AND THE RESULTS OF ALIQUOT CHECKS,
- NAME AND SIGNATURE OF THE PERSON RESPONSIBLE, AND
- MODIFICATIONS OF OPERATING METHODS IF A STANDARD EXISTS.

### Fig. 6 Main Missions Provided by Contract as a Technical Assistance by the CEA-BECC

#### MISSION 1:

Procurements and supplies of technical information and sheets about typical wastes and coatings

#### MISSION 2:

Structure organizing for characterization and expertise planning. BECC equivalent creation.

#### MISSION 3:

Tests and measures by CEA laboratories of real embedded wastes

#### MISSION 4:

Technical training of scientific supervisory personnel for structure organizing and tests of characterizations carrying out

#### MISSION 5:

Setting a quality assurance programme of raw and embedded waste in conformity with each country

# **LOW AND MEDIUM ACTIVITY NUCLEAR WASTE DISPOSAL CHARACTERISATION LABORATORY**

## **Example of Spanish E1 Cabril Disposal Centre Laboratory**

G. Boulanger - Technicatome Espana  
X. Augustin - Technicatome Saclay

### **ABSTRACT**

Low and medium activity radioactive waste generated in Spain by power reactors, research laboratories, etc. is stored in the E1 Cabril Disposal Centre.

This Centre, based on a French design, provides a characterisation function for the stored waste and corresponding containers.

Technicatome, prime contractor for the French disposal centre, and contributing to the design and construction of the E1 Cabril Centre, played an important part in the R&D work for this laboratory, and the manufacture of certain items of equipment.

This laboratory, applying experience acquired in France by the CEA, comprises a set of buildings providing for active and inactive test operations.

### **Introduction**

To ensure that waste generated by the nuclear facilities is compatible with the acceptance criteria established by ENRESA (Radioactive Waste Management Authority) for surface storage, the Spanish Waste Management Programme includes a super-control and characterisation phase, applied to actual waste representative of the various waste production sources.

### **1. Purpose of the Laboratory**

Demonstration of the permanent character of waste disposal packaging is a major source of difficulty. Contemporary modelling of what will happen in tens, hundreds or even thousands of years from now is not a simple matter. The characterisation of a container, demanding precise knowledge of its construction, manufacture, and prediction of its behaviour in the course of time, is the task of the Characterisation Centre. Within the framework of a Franco-Spanish cooperation arrangement, and on the basis of experience acquired with the various phases of the fuel cycle, Technicatome prepared the Basis for Design for the ENRESA (Empresa Nacional de Residuos Radioactivos SA) Characterisation Centre on behalf of CEA/DRDD/SCECA.

### **2. Operations Conducted in the Laboratory**

The objective is to dimension the structures used to characterise the waste containers, and execute super-control operations on these containers. The centre includes the following unites:

- waste characterisation,
- personnel health and monitoring,

- safety of installations and buildings,
- health physics (control of personnel and raw materials),
- infrastructure (maintenance shop, supply and distribution of fluids), and
- local management of the centre.

Inspection and test operations can be conducted on complete containers, or on samples prepared from complete containers, in the characterisation unit.

These containers are active or inactive. Various physical, mechanical, thermal, radiological, and chemical tests for characterisation of the containers and packaging procedures, are conducted according to the nature of the containers and samples.

### **3. General Description of the Facilities**

The facilities comprising the ENRESA characterisation laboratory are integrated in the general infrastructure of the El Cabril Centre. These facilities are located in two separate buildings.

#### **3. 1 Inactive laboratory**

To facilitate its utilisation, the first building is located in the free access zone of the centre. This is the Inactive Laboratory.

These premises are used for two types of activity:

- administrative management of result data for the various tests carried out in both buildings, and
- analyses and tests on inactive samples, reproducing the physico-chemical characteristics only of the waste matrices.

#### **3. 2 Active laboratory**

The second building is located in the controlled zone of the centre, adjacent to the Waste Packaging Shop. This building houses the actual waste super-control facilities.

Application of radiological protection rules and the ALARA criterion, included in the general criteria of the El Cabril Centre, has led to the utilisation of dedicated techniques and resources for the design of the Active Laboratory.

#### **3. 3 Manipulation cell**

The super-control procedure involves waste sample preparation operations, with risks of contamination and irradiation incompatible with the limits set by the presence of personnel. This is why mechanical operations such as stripping of the concrete matrices, obtaining of samples by dry trepanning of the drums, compression, and exudation tests, etc. must be conducted in the shielded containment of a manipulation cell. Handling of these machines and samples, using remote manipulators, is monitored via shielded windows.

Furthermore, distribution of waste or samples to the test equipment and/or for external tests outside the cell, must also be conducted under remote control from a station equipped with man/machine dialogue consoles, assisted by a closed-circuit TV system.

### **4. Active Cell Organisation**

These requirements, combined with the specific nature of the tests to be conducted, led to the

design of the building as described below.

The active building has a total area of 1200 m<sup>2</sup> on two floors.

The ground floor includes a radiological control station for checking personnel, and is also used to control personnel access to the premises. The operator(s) then enter a central equipment control zone, also providing access to the various radiological and radiochemical analysis rooms. One wall of the central control zone forms the front face of the manipulation cell, with two operator stations and four remote manipulators. On one side of this wall, the operator can access a room in which samples extracted from actual drums, or actual stripped drums, which may have been irradiated, are subjected to lixiviation tests, and on the other side, to the room in which waste drums and samples are subjected to spectrometric examination. All handling operations can be remotely controlled from the control consoles mentioned above.

The rear zone of the cell communicates with these premises, and also with the Preparation Shop which supplies the waste samples for examination, without interruption of the general containment of the laboratory.

Auxiliary controlled ventilation equipment and other electrical services are located on the second floor.

### **Conclusions**

Construction of the E1 Cabril Disposal Centre Characterisation Laboratory had the benefit of the extensive experience acquired by Technicatome, resulting from close collaboration with the CEA in connection with a similar laboratory in France.

The largely approved methods applied ensure an excellent level of quality for the containers stored on the disposal site.

## **SESSION V**

### **DESIGN, CONSTRUCTION & OPERATION**

# FRENCH EXPERIENCE IN DESIGN AND CONSTRUCTION OF NEAR-SURFACE DISPOSAL FACILITIES FOR LOW-LEVEL WASTE

JOUSSELIN Daniel      MEDAL Gérard      AUGUSTIN Xavier  
TECHNICATOME, FRANCE  
de WAVRECHIN Bertrand SGN, FRANCE

## ABSTRACT

France disposes of all radioactive waste produced on its territory.

Short-lived waste (with a half-life shorter than 30 years) are disposed of, since 1969 on the "La Manche" disposal facility (CSM "Centre de La Manche").

As this center will be saturated in 1994, ANDRA (French National Agency for Radioactive Waste Management) has undertaken in 1984 the studies and works necessary to the realisation of a new disposal facility.

TECHNICATOME was associated, since the beginning of those studies and was chosen by ANDRA as Prime Contractor for the new Radwaste Disposal Center.

French conception was chosen by Spanish Authorities in 1987, ENRESA (Empresa Nacional de Residuos Radioactivos SA) selected the Cabril Site in the South of Spain as disposal of low and medium activity radwaste. TECHNICATOME was associated with this project, through a joint French-Spanish engineering team.

Authority of North Carolina State (USA) decided in 1989 to build a low-level radioactive waste disposal facility and the contract has been awarded to CNSI (Chem Nuclear System Inc.) with a proposal based on the French experience. A french team ANDRA/TECHNICATOME/SGN is in charge of the design of the disposal facility.

## 1. General

### 1. 1 Management criteria applied to French disposal conception

Design and construction of a near-surface disposal facility for short-lived radioactive waste are based on two main safety criteria :

- waste must be isolated from the environment during the "decay period", i. e. during the time in which the radioactivity decreases naturally to negligible levels; and
- this period, during which institutional control of the disposal facility is necessary, must be limited in time. In France, it was decided that, taking into account the building and operating principles of shallow land disposal centers, this period should not exceed 300 years. The institutional control period could be shortened if at the end of operations it is determined that the total radioactivity of the facility is less than authorized limits.

The first criterion requires that there be no radioactive releases from the facility to the environment under normal operating conditions. In other words, both the waste form and the

disposal structure that receives it must together provide intrinsic safety. To this end, a preventive protection system is deployed in which physical barriers protect the waste's integrity and collect water that might have come in to contact with the waste.

Public health and safety also require that the consequences of an accident be within acceptable limits. Although the probability of such an occurrence is low, it is foreseen that the disposal site itself would act as an additional barrier to the spread of contamination.

The second criterion required limits on the activity levels of waste accepted for disposal, whether this involves short-lived or long-lived radionuclides, such as alpha emitters. Specific limits are determined by the environmental impact study, which includes an analysis of radiological impacts, that is required before permission to construct and to operate the disposal facility is granted.

The safety of the disposal system thus relies upon a certain number of specifications that involve: development of waste forms, disposal facility construction, and site selection.

Once a site has been selected following very specific rules, a waste management system must be implemented to guarantee the quality of the operations carried out on the wastes and structures for a long time. The safety of waste facility depends on the rigor of such a quality control system.

### 1.2 Waste acceptance (French disposal)

Only low- and medium-level waste containing primarily short-lived radionuclides, i. e. less than 30 year half lives, may be accepted for disposal in a near-surface facility.

For each disposal facility, strict rules specify the maximum quantity of each radionuclide allowed in the waste. For example, the following limits are specified in the case of the Centre de La Manche for the most common beta-gamma radionuclides:

Cobalt-60	1,200 Ci/t	44,400 kBq/g
Strontium-90	20 Ci/t	740 kBq/g
Cesium-137	130 Ci/t	4,810 kBq/g

The quantity of long-lived radionuclides must be extremely low so that long-term risks after facility decommissioning are negligible under all circumstances, regardless of the use to which the land is put.

Accordingly, the maximum quantity of alpha emitters must be less than the following: an average of 370 Bq/g for all waste combined and less than 3,700 Bq/g for any individual container of waste.

Furthermore, the waste form must be qualified by ANDRA for acceptability for disposal prior to shipment and must meet ANDRA's specifications.

By waste form, or package, we mean the unit constituted by the waste, which may have been incorporated into a solid matrix and by the container into which it is placed.

There are about 15 types of standard containers for which disposal costs are minimal. However, waste generators are free to choose the method of processing they wish for the waste put into these containers, such as cementation, bituminization, resins, etc. as long as the processing technique has been qualified by ANDRA. A hundred types of waste packages are thus accepted.

When the waste cannot be placed into standard containers, special containers must be used, subject to review and approval by ANDRA and at supplemental cost to the generator.

The sources of disposed waste generally fall into one of the following categories:

- nuclear power reactors, which generate solid waste, air filters, primary coolant water filters, ion exchange resins, and evaporator concentrates;
- spent nuclear fuel reprocessing plants, which generate primarily solid waste acceptable for disposal;
- nuclear fuel cycle facilities; and
- small generators, which produce essentially short-lived beta waste.

The majority of waste is emplaced into one of three broad categories of containers:

- 100 or 200 litre steel drums, which represent the majority of waste received for disposal;
- standardized metal boxes of 2.5, 5 or 10 m<sup>3</sup>; or
- standardized 2 m<sup>3</sup> high-integrity concrete drums, which can contain irradiating waste with higher activity levels up to about one hundred kilo Becquerels per gram; some of waste receipts are in this container.

## 2. The Aube (France) Disposal Facility — Design, Construction, and Operational Startup

### 2.1 Introduction

France produces 80% of its electric power from 59 nuclear power plants. Control of the fuel cycle and long-term control of wastes have been associated to this ambitious program. For example, for nearly 25 years now, France is endowed with a storage centre for low and medium activity nuclear wastes, thus ensuring long-term control of 90% of the volume of wastes produced by its nuclear industry.

The "Manche" disposal facility opened in 1969, was scheduled for closure in 1994 after receiving over 500,000 cubic metres of wastes. It is for this reason that ANDRA decided in 1985 to build a new disposal facility in the Aube Department of France, 200 kilometres from Paris.

It was at this time that ANDRA selected TECHNICATOME for the engineering.

Studies for new French low and medium activity radwaste disposal began in 1984.

TECHNICATOME has performed a set of preliminary studies enabling to determine the main basic data and design options for the facilities to be set up on this new site.

The major items of analysis concerned the optimization of the on-site radwaste packaging and/or treatment modes and the definition of the disposal modes for each type of packages.

The detailed studies undertaken by ANDRA and complementary made by TECHNICATOME enabled to gather all data necessary for the engineering work.

### 2.2 Concise description

The Aube disposal facility is to receive a million cubic metres of low and medium activity wastes during a period of at least 30 years, i.e. 25,000 to 30,000 cubic metres a year. Built on a 100 hectare site, it consists mainly of four parts:

### The disposal zone

This mainly comprises:

- 24 disposal structures (in the complete operating phase, these will be over 400), divided into six groups of 1 to 5 structures. Each group is equipped with a mobile roof and an overhead travelling crane;
- A network of underground galleries beneath each group of structures and on either side of the central track of the site. This gallery contains the Separative Water Collection System (SWCS) for collecting waters that have infiltrated into the loaded structures, the rain water network of the empty structures, and the distribution of power; and
- The terminal structure containing the BGSN water retention tanks and the infiltration waters into the underground gallery, together with the pumping, ventilation, and heating installations.

### Conditioning shops

The 5 and 10 m<sup>3</sup> metal caisson conditioning shop.

Its function is to inject mortar into the caissons so as to dispose them in the structures.

The 200 litre drums conditioning workshop.

This is intended for loading 200 litre drums carried in 20 or 40-foot containers to the 1,000 ton hydraulic press compacting stage, blocking the cakes thus obtained in 400 litre drums using mortar made in the installation. This shop also comprises an automate store for sorting the drums.

### The buildings

- The services building (laboratory, changing rooms, etc.)
- The administrative building (management, administration of the centre, etc)
- The mechanical workshop (servicing of vehicles, etc.)
- Transit building for drums not up to specification
- The gate-house
- The restaurant
- The visitors reception building

### Other technical installations

- The power distribution and transformer substation
- The storm waters reservoir
- The pumping and drinking water station
- The purification station
- The meteorological station
- The helicopter pad

### **2.3 Design**

TECHNICATOME designed the Aube disposal facility on the basis of the following four main objectives:

- Improving the long- and short-term disposal safety;
- Meeting foreseeable changes in regulations, particularly with regard to exposure of the personnel;

- Achieving a satisfactory level of reliability; and
- Obtaining efficient management of the packages.

These objectives were achieved by implementing the following technical solutions.

#### New disposal concept

To prevent the packages from coming into contact with rain waters during the operating phase or runoff waters or possible leakages of the final cover during the surveillance and rehabilitation phases, a new disposal concept has been developed. The disposal structures consist of a foundation pit with sides 25 m long and walls 8 m tall. A metal frame covers the structure during loading, together with part of the adjacent structure containing the transport truck. The packages are handled by an overhead travelling crane of suitable capacity. The space between the packages is filled with gravel or mortar, depending on whether the package has a durable envelope or a metal envelope. The structure is then closed off by a slab and rendered watertight, after which the frame is moved over the adjacent structure. This design offers many advantages: the packages are kept permanently sheltered from the rain when the structure is being loaded and following closure of the structure; once closed, the disposal structures form monolithic blocks providing better behaviour in the presence of seismic shock or distortion of the terrain during the surveillance or rehabilitation phases; the dosage rates absorbed by the personnel are considerably reduced by the existence of two walls between the disposal facility and the zone in which the personnel work; and the cab of the overhead travelling crane is equipped with biological shielding.

#### Automation of the facilities

All the package handling operations on the Aube disposal facility are performed by specialized and remote-controlled equipment.

##### - Storage function

For the four groups of structures used full-time, operation is automatic after pick-up of the packages as regards the various movements, identification of the package, setting down and transmission of the coordinates to the computers.

##### - Drums conditioning unit

This unit, which operates automatically, is controlled from the control room by the control-monitoring supervision and management system.

#### Control-monitoring supervision and management system

The control-monitoring supervision and management system consists mainly of three parts:

##### (1) Management ensuring

- Verification that the packages received at the site correspond to those declared by the generators,
- Preservation of a print of all the operations performed on the packages (compacting, grouting, storage, etc.),
- The ability to pinpoint a package at the site, and
- The drawing up of reports of the activity of the site.

##### (2) Supervision

- For providing the operator with means of control and monitoring of all the conditioning shop

equipment.

- For facilitating maintenance.

- (3) Control and monitoring for
  - the mechanical process,
  - the mortar production installations,
  - the ventilation
  - the power distribution, and
  - managing the active and "dubious" effluent networks.

The control-monitoring supervision and management system consists mainly of two mutually backed up computers, connected by a telematics network to the 19 programmed automats controlling the installations. Five graphic operating stations manage the 60 mimic displays and about 6500 items of information. About 12 alphanumeric consoles and printers ensure management and supervision. The control-monitoring supervision and management system is connected by TRANSPAC to the main-frame computer at the ANDRA head office.

The same design rules as for reactors

The conditioning units of the caissons and drums have been built applying the same rules as those governing the design of nuclear reactors, in particular with regard to nuclear safety, zoning, confinement, ventilation, radiological protection, detection means, and firefighting means.

#### 2. 4 Construction

The work of constructing the Aube disposal facility took place over a 5-year period. Nonetheless, two main phases can be distinguished:

- General preparation of the site, construction of the disposal structures, and caisson conditioning units

1987 - Preparation of the site, construction of the access road to the centre, and clearance of growth

1988 - General earthmoving and execution of the storm waters reservoir

1989 and 1990 - Execution of roads and various networks

From July 1989 to February 1990 - Construction of the disposal structures, SWCS, and terminal structure

From January 1989 to January 1991 - Construction, erection/installation, and testing of the mobile roofs and overhead travelling cranes

From July 1989 to May 1991 - Construction, erection/installation, and testing of the caissons conditioning unit

- Construction of the drums conditioning unit

The construction, erection/installation, and testing of all the equipment for this unit took place from January 1990 to September 1992.

#### 2. 5 Operational startup

The transfer of the installation for operation occurred between ANDRA and TECHNICATOME from 1991 to Sept. 1992 at full satisfaction of ANDRA the owner.

The Aube disposal facility has been running since the 13th January 1992.

After 13 months of operation, over 8,000 packages have been stored (as of end February 1993).

The drums conditioning unit has been operating since the 25th January 1993.

## 2.6 Conclusion

Constantly striving to possess a nuclear industry that generates no pollution, France has built and operated since the beginning of 1992 a modern, high performance wastes disposal facility. Applying novel, reliable, and safe technical solution combined with wide dissemination of information to the public, this achievement has resulted in acceptance of this low and medium activity wastes disposal facility by the population. Under the pressure of the French Ecologists Lobby (about 20% of votes), the Authorities are gradually requiring the same constraints as for other industries for wastes.

## 3. Spain Low and Medium Activity Radwaste Disposal: El Cabril Center

The radioactive waste general management plan presented by ENRESA (Empresa Nacional de Residuos Radioactivos S. A.) to the Ministry of Industry and Energy, and approved by the Spanish Government in October 1987, led ENRESA to undertake the studies and work required for the construction of a low and medium activity Radwaste Disposal Facility.

The selected Cabril site is currently being used by ENRESA for temporary storage of LA and MA Waste while awaiting final disposal. The site is located in the Sierra Albarrama in the province of Cordoba.

To achieve the objective set by the Spanish Government, namely to have the disposal facility operational by 1992 ENRESA decided to associate the following companies with the project:

- INITEC, a Spanish engineering company, and
- TECHNICATOME, a French engineering company, responsible for construction of the second French Radwaste Disposal Facility in the department of Aube.

The creation of a joint French-Spanish engineering team, makes it possible to adopt safety and design principles already applied in France, with respect for final near-surface disposal of LA and MA radioactive waste, for the Cabril project.

### 3.1 Cabril site

This site has a total area of 1200 hectares, and is located in the Sierra Albarrama (Sierra Morena Massif) in the Province of Cordoba, 450 km away from Madrid to the south.

The selected zone has an area of about 30 hectares, and comprises, in geological terms, a folded series of crystalline schists, gneiss, and mica schists. The fracture density is low.

### 3.2 General organization of the site

All wastes arriving on the site in 200 l metallic drums are placed inside concrete containers in which they are blocked by mortar so that to make a concrete block weighing 24 tons with external dimensions 2.25m×2.25m×2.20m.

Then these containers are stored in works like platforms. The modular design of these works and the standardization of the waste container enable a remote operation.

Prior to the disposal the waste conditioning workshop set up at El Cabril allows a volume reduction by compaction of technological waste by incineration of waste originated by small generators, and packaging of secondary wastes (solids and liquids) generated by the center's facility.

### **3.3 Disposal platforms**

The design principles of the disposal platforms aim to isolate waste containers from water and to limit the radioactivity with respect to the workers and the public.

#### **3.3.1 Platforms**

Each platform is constituted by a concrete flooring which received the containers and sustaining the concrete walls.

External dimensions are 24m × 19m × 10m each platform being able to dispose 320 concrete containers.

During the operation phase, the work is over topped by a mobile roof ensuring protection against rainfalls. This roof includes handling means for concrete containers with remote operation.

The lower concrete flooring is made of different waterproof or porous layers enabling to collect and drain the potential seepage towards the underground collector.

After laying down the containers in the disposal vault concrete roof is placed to close the work. This roof receives a temporary cover waiting for the final one.

#### **3.3.2 Final coverage of the structure**

The disposal structures, for which the operational phase has been completed, are protected against the ingress of rainwater by covering with impermeable, self-draining material, combining with the raft foundation to form the second containment. The waterproofing system of this cover restricts infiltration to a minimum.

#### **3.3.3 Collection and monitoring of infiltration water**

All structures are equipped with systems for the collection and monitoring of any infiltration water passing through the final coverage.

This underground system makes it possible to monitor the first containment system in its entirety, and part of the second containment system (final coverage) of each disposal cell. The design of this system makes it possible to identify the point of degradation in a disposal structure, if the presence of contaminated infiltration water is detected. The system is designed to operate throughout the surveillance phase.

### **3.4 Waste embedding shop**

The waste processing and embedding operations are conducted on the site in dedicated premises designated the "Waste Embedding Workshop".

The three main purposes of this shop are:

- compaction of compactable waste,
- embedding of waste from small producers, and
- treatment of waste produced in the center.

The quantities processed per year should be approximately as follows:

- 8,000 steel drums for compaction,
- 6,000 litres of scintillation liquid for incineration, and
- 2 tonnes of biological waste for incineration.

The initial operation is to receive, off-load and place in temporary storage, using appropriate handling equipment, the waste delivered for embedding in containers or trailers.

Technological waste placed in 200-litre steel drums is compacted with a 1000-tonne press, on a batch basis.

Scintillation liquid and incinerable solid waste is then incinerated on a batch basis.

All treated waste resulting from the compaction operation is blocked in concrete containers by injection of a hydraulic binder.

The majority of waste compaction, incineration, and blocking operations are fully automated.

The remote control equipment and radioprotection data panels are grouped in the embedding shop control room.

### 3. 5 Conclusion

Conjugated actions conducted by the Employer ENRESA on the one hand, and the joint French-Spanish engineering team comprising TECHNICATOME and INITEC on the other, will make it possible to construct the Cabril Radwaste Storage Center and commission the Center in 1992, achieving these objectives and meeting the various safety regulations and requirements concerning the final disposal of LA and MA waste.

## 4. North Carolina Low-Level Waste Disposal Facility

The Low-Level Radioactive Waste Policy Act, passed by Congress in 1980, and the Low-Level Radioactive Waste Policy Amendments Act of 1985 require states to provide for the timely disposal of low-level radioactive wastes (LLRW) generated within their borders. In response to this legislation, Alabama, Florida, Georgia, Mississippi, North Carolina, South Carolina, Tennessee, and Virginia joined together to form the Southeast Interstate Low-Level Radioactive Waste Management Compact (Compact). South Carolina, the first host state for the Compact, is currently providing LLRW disposal service for the Compact at the Barnwell Waste Management Facility Site (Barnwell). Barnwell is operated by Chem-Nuclear Systems, Inc. (CNSI). North Carolina will serve as the second host state.

CNSI has developed the conceptual design for the North Carolina Low-Level Radioactive Waste Disposal Facility (Facility) based on the French concept in association with ANDRA-TECHNICATOME and SGN, grouped as "French Engineering Group".

### 4. 1 Facility summary

The Facility will be located within one of two potential suitable sites. The final site will be selected after site characterization programs when each potential suitable site has been completed and evaluated.

#### 4. 1. 1 Facility layout

The Facility will be divided into two distinct areas:

- the restricted area, which will include most of the waste receiving, handling, packaging, and disposal facilities; and
- the unrestricted area, which surrounds the restricted area and includes the buffer zone.

The restricted area, which will be surrounded by a security fence, will include:

- disposal units,
- Waste packaging building,
- Waste package storage building,
- Movable building,
- overpack storage area,
- access roads, and
- disposal area storm runoff and drainage control structures.

The unrestricted area will include:

- Administration Building,
- Site Access Building,
- Maintenance Building,
- Truck Inspection Building,
- Warehouse Building,
- Utility Building,
- electrical supply, firewater tank, sewage treatment plant, and other utility systems,
- fuel dispenser,
- access roads,
- parking areas, and
- storm runoff and drainage control structures.

#### **4.1.2 Design overview and waste packaging**

The French concept and experience is used for this disposal.

The Facility design incorporates the use of the Integrated Vault Technology (IVT) proposed by CNSI. The IVT consists of above-grade concrete disposal units, made up of reinforced concrete disposal modules which will be filled with waste packages and covered by an engineered earthen cover.

Infiltration of water into the disposal modules is not anticipated; however, for added protection, an infiltration collection and detection (ICD) system will be installed within each disposal unit. This monitoring system will detect infiltrated water draining from the disposal modules and sound an alarm if water is detected. Detection equipment will be fully accessible for repair, replacement, or upgrade as technology advances during the operating life of the Facility.

After the active disposal module is fully loaded, a reinforced concrete roof slab will be cast in place to cover and seal the disposal module. The Movable Building will then be moved to cover the next active disposal module. The filled disposal modules will be covered with an engineered earthen cover and monitored to demonstrate compliance with regulatory requirements.

#### **4.2 Assumptions**

The following assumptions were used in the development of the DBS:

- the disposal modules will contain Class A, Class B, and Class C LLRW;
- the Facility will operate for a period of 20 years;

- North Carolina General Statute requires that the state of North Carolina provides disposal capacity for 20 years or 32 million cubic feet of waste, whichever occurs first. The two potentially suitable sites currently being characterized will provide the capacity for 32 million cubic feet. However, current projections of waste inventory show that only approximately 11 million cubic feet will require disposal during the 20-year operation period;
- disposal operations will be scheduled primarily to occur during one 8-hour week; supplemental shifts will be scheduled if necessary;
- disposal module construction will require excavation of unsuitable surficial soils. Excavated areas will be backfilled with selected materials prior to construction;
- the bottom of the Facility as it relates to North Carolina General Statute is the bottom of the waste package which coincides with the finished floor elevation of the disposal module; and
- waste container sizes, shapes, and features will be similar to those presently used by generators.

## 5. Other International References

The French surface disposal design for low and medium activity short-life radioactive waste is now recognized internationally.

The basic principles are unchanged, only modifications have been made to equipment construction and to operating and supervisory procedures, thus drawing on the benefit of more than 20 years experience in the operation of the Manche Disposal Facility and the construction and commissioning of the Aube Disposal Facility.

The skills of ANDRA and its engineering working in this area via TECHNICATOME and SGN were put to optimum use internationally as part of cooperation agreements confirmed by many advisory and evaluation assignments.

After Spain, where TECHNICATOME was fully involved with a Spanish engineering company, the French concept has henceforward been adopted by several countries among which are several "compacts" in the United States, Japan and more recently in Mexico. These countries asked for French assistance to study the feasibility, design, and even engineering of their surface disposal on the basis of the principles applied by ANDRA in France.

### 5.1 Prime contracting assistance assignment

The French participation in international disposal projects first led to consulting assignments including primarily:

- Oak Ridge, Tennessee (USA)

Working with ANDRA, in 1987, SGN drew up a full report for the Department of Energy (DOE) concerning the French experience in terms of surface disposal.

This study groups together regulation requirements, choices, and qualifications of sites, waste acceptance criteria, principles of disposal design, etc.

The report was followed by a case study carried out on the basis of typical radioactive waste packages existing in the USA and an operator training program for DOE personnel in France.

- State of Illinois (USA)

The IDNS (Illinois Department of Nuclear Safety), as part of the engineering package for its disposal center, called in SGN/ANDRA to define the basis of its computerized waste package tracking system, similar to the French system developed by ANDRA and running on the CSM and CSA disposal centers.

A conceptual study was then carried out on the principles of disposal safety applicable in Illinois. They cover four main subjects:

- Waste acceptance criteria and source term,
- Boundary conditions for safety analysis,
- Concrete degradation model, and
- Structural performances of concrete.

- Japan

In cooperation with ANDRA, SGN drew up a full report on the radioactive waste package tracking system more particularly package traceability from generation at generators through to final disposal.

SGN then developed a computerized tracking system for FEPC (Japanese Federation of Electrical Power Companies) and gave a demonstration at the clients in Japan.

The operator manual drafted at this time takes into account the regulation and particularities of the Japanese organization for radioactive waste management.

Finally, SGN, working jointly with ANDRA, studied the characterization of Japanese radioactive waste, illustrated by case studies on typical waste produced by Japanese plants.

This study was the practical application to the Japanese case of the management principles and methods developed by ANDRA as used in France.

- Taiwan

In 1989, SGN and ANDRA carried out a mission to evaluate the management practice used for radioactive waste in Taiwan. The mission was completed by the publication of a report including recommendations for the establishing of a general radioactive waste management program and defining safety objectives for the management and disposal of the waste.

### 5.2 Participation in the production of surface disposal

The international participation of the French, in addition to the above described Spanish example applied to production studies among which we might mention :

- Mexico

With its Eurisys network, SGN and ANDRA were engaged in a feasibility study for surface disposal of radioactive waste on the Laguna Verde site.

This study is a site survey with evaluation of its characterization, an inventory of the waste packages to be disposal, the design of the disposal on the CSA model and the definition of the principles and safety options.

### 5.3 NPP on-site disposals

Some countries have been thinking of on-site disposal facilities for 2 or 4 units. These disposals have to be designed as a Monitoring Retrievable System (MRS). TECHNICATOME taking

advantage of its experience has made feasibility studies of such facilities located close by the NPP.

#### 5.4 Conclusions

Thanks to their experience in France and abroad in the field of low to medium activity short-life waste surface disposal, ANDRA, SGN, and TECHNICATOME are capable of applying the waste management principles defined by ANDRA to the design and production of disposal facilities having different characteristics according to the chosen country or site while guaranteeing an optimum level of safety during operation and after shutdown.

In close conjunction with ANDRA, TECHNICATOME and SGN have demonstrated that they are capable of working together with the government organization agents, local authorities, and the public concerned in order to take into consideration their demands or requirements at the design stage and through to the operating procedures used within the centers.

# **ASSUMPTION AND PROGRAM OF THE EARLIER STAGE CONSTRUCTION OF L/ILW DISPOSAL SITE**

LI Xuequn

CHEN Shi LI Xinbang

China National Nuclear Corporation

China Institute for Radiation Protection

## **ABSTRACT**

This paper analyses the production and treatment of low- and intermediate-level radwastes (L/ILW) in China, and predicts these wastes to be produced while developing nuclear power. Then it introduces some problems and situation in this field and puts forward some proposals and assumptions to the earlier stage construction of disposal site.

With the development of nuclear industry, quite a large amount of low- and intermediate-level solid radwastes have been accumulated, which has become a safety-related problem increasingly important to nuclear enterprises and institutions. More and more such wastes are being produced with the nuclear power development and nuclear facilities decommissioning. Because the wastes must be disposed of safely, it is absolutely necessary to construct the disposal sites. Therefore, preliminary efforts have been made by CNNC over the past ten years in policy, law and rules, developing program, management system, siting, engineering techniques, and safety assessment for waste disposal. However, due to lack of definite technology policy and law, management organization, and construction funds, this work remains in the stage of academic research, policy deliberation, and siting study, which can not meet the requirement of radwaste disposal.

The present situation and prediction of the L/ILW, and the program and assumption to disposal site construction are reviewed as follows.

### **1. The Status Quo and Prediction of Low- and Intermediate-Level Solid Radwastes**

50,000 m<sup>3</sup> low- and intermediate-level solid radwastes, including solidified bodies of evaporated liquid wastes, have been accumulated in China over the past 30 years. In the next decade, the same quantity of wastes will be generated with the nuclear facilities decommissioning.

Based on nuclear power development program in the near future, one 1,000 MWe standard light water reactor produces 550 m<sup>3</sup> low- and intermediate-level solid radwastes each year in terms of international common estimation and parameters; and 40 years later, there will be the same amount of the wastes to be generated when it is out of service. So, the amount of such solid wastes will amount to 380,000 m<sup>3</sup>. The wastes will increase with the development of nuclear power. Produced by nuclear industry as well as nuclear power plants, hundreds of thousands m<sup>3</sup> of solid wastes are expected for treatment.

In the past, the disposal of L/ILW was not dealt with in time, especially those wastes which have been stored in the temporary storage pits for a long time and it is now difficult to retrieve them. Due to the time limit of the construction and in case of rainwater leakage in the constructions, the radionuclides will be released and migrate. When new nuclear power plants and nuclear facilities are put into operation, the waste disposal shall be taken into consideration.

It should be stipulated that the temporary storage of low- and intermediate-level solid radwastes by the site area is limited within 5 years as a necessary measure to ensure safety. Consequently, the construction scale and standard of the temporary storage can be optimized, and economic benefits, effected as well.

Authorities concerned in CNNC have concluded unanimously that the construction of the L/ILW disposal site for nuclear facilities shall be regarded as a prerequisite for development of nuclear power and decommissioning of nuclear facilities and as an important content for the examination of safety analysis and environmental impact assessment of nuclear facilities by the environmental protection and safety supervision authorities.

## 2. Assumption and Program of L/ILW Disposal Site

L/ILW disposal site is not only used in nuclear power plants and nuclear industry, but also can facilitate the development and application of radioisotopes, irradiation technology, and other nuclear technologies. According to the principle of regional disposal and distribution of the L/ILW, five regional disposal sites are to be constructed in the east, south, northwest, southwest, and northeast of China in the present and intermediate periods. With an exception of the northeast disposal site which can be postponed for construction, the first phase engineering of the others is scheduled to be finished and put into operation by 2000. If the above objectives are realized, the requirements to dispose of the wastes arising from nuclear industry, nuclear power plants, nuclear facility decommissioning, etc. can be met. The estimation of the scale and costs of those five sites is listed in Table 1. The investment is made by stages of construction and application of the site.

There is an obvious difference in the construction procedures between a disposal site and a general nuclear facility. The waste disposal means to bury a certain amount of radioactive materials underground with support of geological and engineering barriers and other management measures so as to ensure safety for 500 years until the radioactive materials decay to be harmless products. Therefore, in pre-construction stage, it takes a long time to select an appropriate site, perform safety assessment, and implement the official review and approval. The construction cycle in foreign countries usually takes 5~7 years, including 4~5 years long for siting and safety assessment. Large amount of work in siting involves field investigation, drilling, measurements, and in-situ tests; geological surveys such as hydrogeology and geochemistry are more detailed than those for siting of a nuclear power plant. The above conditions are of great importance to the earlier stage work of site construction. Mainly because of financial difficulty, the construction of such site can not proceed steadily.

## 3. Earlier Stage Work and Proposed Work during the Eighth-Five-Year Plan

The earlier stage work includes the selection of technology, regional investigation, preliminary site selection, site determination, safety tests, safety assessment, conceptual design, and preliminary design. The earlier stage work and specific conditions of the site are closely interrelated. The earlier stage work includes the following technical items:

- (1) Investigate the geological, hydrogeological, environmental, ecological, and social economic conditions in each region, and recommend several possible areas;
- (2) Carry out reconnaissance in these possible areas and select over 20, and then to recommend

- one or more possible sites on the basis of comprehensive assessment and drilling tests;
- (3) Implement conceptual design of the waste disposal system according to specific conditions of the site;
- (4) Prepare feasibility research report, safety analysis report in siting stage, and environmental impact report;
- (5) Assess in detail the characteristics of one or two examined and approved sites, including a. Ground survey and drilling in field; b. Determination of the field hydrogeological and engineering geological parameters such as water pumping and pouring tests, ground water tracer tests, single well dilution tests, unsaturated zone tests, geophysical exploration and the age of fracture movement; c. Determination of hydrogeological and engineering geological parameters in laboratory such as rock and soil porosity, permeability coefficients, grain size, specific gravity, the characteristics of rock core samples in different depths, rock and soil mineral and chemical composition, water quality analysis of surface water and ground water, radioactivity background survey of ground water, radioactive nuclides migration tests, geochemistry research, the age of ground water, and rock and soil mechanics;
- (6) Determine the disposal methodology, engineering techniques, and engineering barrier parameters, including a. The shallow ground disposal and rock-cave disposal; and b. Leaching tests, corrosion-resistant tests of packing container, backfilling materials tests, top cover tests, and the structure and material tests of other types of disposal unit;
- (7) Study the safty assessment techniques of a specific disposal system, including a. Suitability R&D of the assessment modes and computer programs, especially the integral mode, failure mode of engineering barrier, aerated-zone radioactive nuclides migration mode and moisture content movement mode, ground water radioactive nuclides migration and water content movement mode, and probability risk analysis and evaluation; b. Uncertainty analysis of assessment mode, sensitivity analysis, and verification of mode; and c. Determination of key radioactive nuclides, key routes, and key residential areas, and ecological investigation of food-chain and determination of transfer parameters;
- (8) Prepare safety analysis report and environmental impact report on the basis of preliminary design; and
- (9) Ensure the quality of the earlier stage work, including preparation and implementation of QA Program.

Since most of the above work aims at specific site and disposal system, all the requirements must be met in all the regional disposal sites. As proposed, the siting and other primary earlier stage work in the east, south, northwest, and southwest of China should be finished during the Eighth-Five- Year Plan to lay a solid foundation for the first phase engineering of the four disposal sites by 2000.

Regional differences in cost and existing work conditions are considered in the investment. The cost estimation is listed in Table 2(investment for the preliminary design excluded).

In order to avoid building all the disposal sites at the same time to disperse the technical capabilities, the eagerness, siting conditions, and the supporting work by related departments in each disposal region should be taken into account when the earlier stage work is arranged in each disposal site. The proposed time schedule is shown in Table 2.

In recent years, in respect of low- and intermediate-level liquid radwastes disposal, CNNC has been involved in two earlier stage works, i. e. near-surface disposal for cement solidified wastes

by large-volume pouring technology and the hydraulic fracturing disposal in the shale layer, reaching an extent of determination of the disposal sites. And the preliminary siting stage work in the east, northwest and southwest of China has begun. Through practical work, experience has been gained, and a technical contingent, built up.

In addition, cooperation has been developed with several foreign countries and institutions in L/ILW disposal with fruitful results. All those factors are considered as the advantages to finalize the L/ILW disposal sites during the Eighth-Five-Year Plan.

The L/ILW disposal sites shall be constructed with supports to be given by related departments to speed up the progress and build up the sites as early as possible so that the development of nuclear power and nuclear industry and application of nuclear techniques can be ensured in China.

Tabel 1 Estimation for construction of the disposal sites

Disposal Site	Effective Volume * (10 <sup>4</sup> m <sup>3</sup> )	Investment (RMB¥1,000.00)	Investment by 2000 (RMB¥1,000.00)
East China	20	50,000	30,000
South China	20	50,000	30,000
Northwest China	20	30,000	20,000
Southwest China	5	20,000	15,000
Northeast China	20	50,000	
<b>Total</b>		<b>200,000</b>	<b>95,000</b>

\* According to actual conditions, the construction is carried out by stages.

Table 2 The investment of the earlier stage work of L/ILW disposal site construction during the Eighth-Five-Year Plan

Disposal Site	Investment (RMB¥1,000.00)	Duration
East China	2,500	1993~1995
South China	3,000	1991~1994
Northwest China	1,500	1992~1993
Southwest China	2,000	1993~1995
<b>Total</b>	<b>9,000</b>	

**SESSION VI**

**ENVIRONMENTAL, SAFETY MONITORING**

# NORTHWEST DISPOSAL SITE FOR LLW AND ILW IN CHINA RADIOACTIVE IMPACT ASSESSMENT

WEI Kuizi HE Chunying LU Baozhen LI Tingjun

Beijing Institute of Nuclear Engineering, China

## ABSTRACT

This paper describes the studies and main conclusions in site selection, design, and radioactive impact assessment of the Northwest Disposal Site for intermediate- and low-level radioactive wastes. At the end of the paper, further works are proposed.

### 1. Introduction

The Northwest Disposal Site as one of the four proposed regional disposal sites for intermediate- and low-level radioactive wastes in China is located in a dry, vast, and sparsely-populated area, and it is close to the Lanzhou Nuclear Fuel Reprocessing Plant in Northwest China. The construction of the site will not only benefit to solve the problem of the wastes generated from the Lanzhou Nuclear Fuel Reprocessing Plant, but also provide experience for construction of other disposal sites in China.

### 2. Site Selection

Through the previous regional investigations, two sites have been selected for final determination.

The two candidate sites, 7 km away from each other, are located in the Gobi Desert.

#### 2.1 Site characteristics

The investigation of geological, seismic, hydrological, and hydrogeological characteristics, natural and social environment of the sites has been completed. Table 1 shows the main results of the investigation.

Fig. 1 shows the formation of subsurface layers underneath the sites.

#### 2.2 Site evaluation

The main advantages include:

- a. High stability in regional geological structure, no active faults;
- b. Low precipitation, high evaporation, low population density, no exploitable resources;
- c. Weak seismic activity;
- d. No poor engineering geological conditions such as landslide, avalanche, liquidation of sandy soil, etc.;
- e. Great depth to groundwater, thick clay layer with low permeability in the vadose, strong retardation to nuclides of Sr-90, Cs-137, etc. ; and
- f. Far away from surface water, no flood risk.

The main disadvantages are as follows:

It is perennially windy and the maximum wind speed reaches 28 m/s, and there is risk of slow erosion to the overburden of disposal units.

The two sites are compared in Table 2.

The characteristics of both sites meet the requirements of the national standard (GB 9132) for LLW & ILW disposal, and site 2 is better.

### 3. The Description of Wastes

The wastes to be disposed of in the site are mainly the intermediate- and low-level wastes arising from Lanzhou Nuclear Fuel Reprocessing Plant at present and by 2005, and also those generated in other areas of the Northwest China.

#### 3. 1 The specific activity of the wastes

According to the national standard GB 9133, "The Standard for Classification of Radioactive Waste", the wastes to be disposed of include:

- a. The waste with the radionuclides of half-life  $\leq 60$  d, specific activity (SA)  $> 7.4 \times 10^4$  Bq/kg and  $\leq 3.7 \times 10^7$  Bq/kg is low level radwaste (LLW), and the waste with SA  $> 3.7 \times 10^7$  Bq/kg and  $\leq 3.7 \times 10^{11}$  Bq/kg is intermediate-level radwaste (ILW);
- b. The waste with the radionuclides of half-life  $> 60$  d and  $\leq 5$  a (including Co-60), with SA  $> 7.4 \times 10^4$  Bq/kg and  $\leq 3.7 \times 10^6$  Bq/kg is LLW, and the waste with SA  $> 3.7 \times 10^6$  Bq/kg and  $\leq 3.7 \times 10^{11}$  Bq/kg is ILW;
- c. The waste with the radionuclides of half-life  $> 5$  a and  $\leq 30$  a (including Cs-137), SA  $> 7.4 \times 10^4$  Bq/kg and  $\leq 3.7 \times 10^6$  Bq/kg is LLW, and the waste with SA  $> 3.7 \times 10^6$  Bq/kg and  $\leq 3.7 \times 10^{10}$  Bq/kg is ILW; and
- d. The waste with the radionuclides of half-life  $> 30$  a, SA  $> 7.4 \times 10^4$  Bq/kg and  $\leq 3.7 \times 10^6$  Bq/kg is LLW, and the waste with SA  $> 3.7 \times 10^6$  Bq/kg and  $\leq 3.7 \times 10^9$  Bq/kg is ILW.

#### 3. 2 Types of the wastes

There are two types of the wastes, i. e. the metallic and the non-metallic wastes. The metallic wastes include abandoned pipes, valves, filters, etc. and the non-metallic wastes include graphitic casing pipes, plastic surface covers, work clothes, etc.

The radionuclides mainly consist of Sr-90 and Cs-137.

#### 3. 3 Packaging of the waste

The carbon steel drums of 200 l, 610 mm in outer diameter and 910 mm in height are used for the low-level waste.

Two types of reinforced concrete drums are used to pack the intermediate-level waste. Both have a diameter of 1400 mm and a height of 1300 mm, but the wall of one type is 400 mm thick and the other 300 mm. The maximum volume of the former is 400 l, and of the latter, 200 l.

The allowable levels of radioactive contamination on the drum surface are:  $\alpha < 0.4$  Bq/cm<sup>2</sup> and  $\beta < 4$  Bq/cm<sup>2</sup>. The surface dose equivalent rate of each drum shall not exceed 2 mSv/h.

## 4. Design of the Disposal Site

### 4.1 Capacity

According to the volume of the waste drums, the disposal capacity of the disposal site to be built is  $60,000 \text{ m}^3$ , and the gross radioactivity to be disposed of is  $1.24 \times 10^{16} \text{ Bq}$ . The planned capacity is  $200,000 \text{ m}^3$  and the planned gross radioactivity to be disposed of is  $4.13 \times 10^{16} \text{ Bq}$ .  $5,000 \sim 10,000 \text{ m}^3$  of wastes will be received per year.

### 4.2 Procedure

When the waste packages arrive at the disposal site, the surface dose rate, surface contamination level, and the integrity of the packages shall be checked first. The qualified packages will be registered and then placed in the disposal units or the interim-store room, depending on the operation conditions. The unqualified, if its property or specific activity is inconsistent with the stipulation of the disposal site, its package is damaged, or its contents greatly deviate from those defined in the list, will be rejected. If the waste packages are only damaged lightly or the surface contamination only slightly exceeds the standards, they can be repacked or decontaminated on the disposal site, and rechecked. The disposal procedure is shown in Fig. 2.

### 4.3 Area division

The disposal site is divided into two areas, controlled area and uncontrolled area.

The controlled area consists of disposal zone, buffer zone, and operation zone. The disposal units are placed in the disposal zone. Between the disposal zone and the outside environment is an isolated zone called buffer zone where some monitoring points are set up. The wastes are checked, registered, repacked, and decontaminated in the operation zone.

The uncontrolled area consists of the administration zone and public service zone.

Fig. 3 shows the planning diagram of site division, and the occupied area and the construction area of each zone of the disposal site is shown in Table 3.

Fig. 4 shows the areas, zones and the facilities of the disposal site.

In order to restrict the entry of non-operators, there is a barbed-wire fence around the buffer zone.

### 4.4 Burial scheme

At the site, the groundwater is deep in the stratum, the strong wind exists all the year, and the vegetation is scarce, therefore, near surface trench burial is adopted so as to decrease the erosion of wind, sands, and rain.

There are 9 trenches in the disposal zone and 6 disposal units in each trench (see Fig. 5).

### 4.5 Disposal units

There are two design schemes to be considered for the disposal units.

For scheme 1, there are reinforced concrete channels with reinforced concrete top but no bottom slabs. See Fig. 6.

For scheme 2, there are reinforced concrete channels with reinforced concrete top and bottom slabs. See Fig. 7.

To save construction materials, each unit is divided into four sub-units. The sub-unit is  $14.5 \times 7.5 \times 4.5$  m in internal dimension. Four layers of waste drums can be emplaced, each layer placing 50 intermediate-level waste drums or 828 low-level waste drums. The intermediate-level waste drums must be emplaced at the bottom layer so that the low-level waste drums at the upper layers can play a role of shielding. When a disposal unit is full with waste drums, the space between the waste drums, and the space between the drums and the concrete wall are filled with soil, and then the concrete is poured to form a top slab.

#### 4.6 Drainage system

The open drainage ditches in the disposal zone are constructed to discharge rain water and the surface water entering the disposal units during operation. The slope of the ground surface in the disposal zone is 2%.

The drainage system at the bottom of the disposal units is constructed to discharge rain water that enters the disposal units during operation.

In scheme 1, three transverse drains, a longitudinal drain, and a water collecting pool are set at the bottom of each sub-unit. Rain water flows through the transverse drain to the longitudinal drain and the water collecting pool, then it is pumped into the surface open drainage ditches through the drainage pipes as shown in Fig. 8. The bottom slope of the unit is 1%.

In scheme 2, the main drainage pipes are embedded in the bottom slabs. Each sub-unit is set with by-pass pipes which are connected to the main pipes. At each disposal trench end there is a water collecting pool. Rain water in the water pools is pumped into the surface open ditch through the pipes. The bottom slope of the disposal unit is 1% (see Fig. 9).

When the disposal site is closed, some of the water pipes in scheme 1 and all the water pipes in scheme 2 will be used as monitoring pipes. These monitoring pipes must be well controlled to prevent the surface water from entering the disposal units through them.

#### 4.7 Overburden layer

In the design of the disposal unit overburden layer, the potential erosion, biointrusion, and seepage are considered. The overburden layer designed is 2.5 m thick with 6 layers, which are, from bottom to top, the backfilling soil, pressed clay, bitumen film, gravel or fine sands, pressed clay and cobble, as shown in Fig. 10.

The multiple layer structure design of the overburden layer is to reduce the water entering the disposal unit to the minimum. The top cobbles are designed to protect against wind erosion and biointrusion. The gravel and fine sand layer above the low permeable clay and bitumen film is used as a channelling layer to drain the seepage water into the two sides of the disposal trenches and prevent its further permeating. When a small amount of water permeates through the bitumen film and clay layer, it can seep into the soil layer under the disposal units through the backfilling soil and will not enter the disposal units. The bitumen film is placed below the maximum depth of frozen soil to avoid the effects of freezing and thawing.

## 5. Assessment of the Radiation Impact

### 5.1 Pathways of radiation impact

The possible pathways of radiation impact from the waste disposed of in the Northwest Disposal Site to the public are shown in Figure 11.

### 5.2 The radiation impact in case of unexpected intrusion of the public

After the closure of the disposal site, it is possible that the repository will suffer intrusions as a result of human activity (such as excavation). To predict the radiation dose to the intruders, it is hypothesized that:

- a. the disposal units are semi-infinite volume source; and
- b. the covering soil is completely taken away so that the concrete trough is thoroughly exposed.

If the self absorption is considered as the only way of exposure and the multi-scattering is ignored, the dose equivalent rate of a point outside the semi-infinite volume source is calculated as follows:

$$H = \frac{2\pi \cdot S_v \cdot K_\gamma}{\mu_s} K \cdot E (\mu \cdot t)$$

where

$H$  : dose equivalent rate (rem/h)

$S_v$  : the specific activity (Ci/cm<sup>3</sup>) of volume source

$K_\gamma$  : gamma radiation rate constant (R · m<sup>2</sup>/h · Ci)

$\mu_s$  : the real absorption coefficient of volume source (1/m)

$K$  : the transformation factor of unit (rem/R)

$E(\mu \cdot t)$  : E function

$\mu$  : the liner weakening factor of shielding material (1/cm)

$t$  : the thickness of shielding material (cm)

According to the above calculation, the dose equivalent rate to an unexpected intruder in the disposal area is  $6.96 \times 10^{-8}$  Sv/h.

Therefore, the dose equivalent rate to an intruder is very low.

### 5.3 Assessment of the impact of radionuclides release

During the normal operation and the effective life of the engineered barriers, the safety of disposal units will not be affected by surface and ground water. But in case of certain accident or failure of engineered barrier, the rain water will probably infiltrate into the disposal units and result in release of radionuclides into groundwater. The released radionuclides are then transported to the surface water system through the vadose and the phreatic layer, resulting in exposure on the members of the public.

#### 5.3.1 Source items

It is assumed that the water soaks into the intermediate-level radioactive waste drums at the bottom of the disposal units.

The release rate of radionuclides to groundwater is

$$P_i = \frac{S}{V} R_i \cdot A_i$$

where

$P_i$  : the release rate of radionuclide  $i$  to groundwater (Bq/d)

$S$  : the geometric surface area of waste ( $\text{cm}^2$ )

$V$  : the geometric volume of waste ( $\text{cm}^3$ )

$R_i$  : the leaching yield of radionuclides  $i$  (cm/d)

$$R_{Cs} = 1 \times 10^{-2} \text{ cm/d}$$

$$R_{Sr} = 1 \times 10^{-3} \text{ cm/d}$$

$A_i$  : the gross activity of radionuclides  $i$  (Bq)

### 5. 3. 2 Mode of calculation

#### a. The transport law of radionuclides in the unsaturated zone

When it is assumed that the stratum medium is porous and homogeneous, the bulk density and the effective porosity are steady, and there is a seepage in vadose, then the one dimensional equation of radionuclides transport in porous medium is

$$D_x \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} - \lambda \cdot C \cdot R_d = R_d \frac{\partial C}{\partial t}$$

Where

$C$  : the concentration of radionuclide ( $\text{Bq}/\text{m}^3$ )

$D_x$  : dispersion coefficient ( $\text{m}^2/\text{s}$ )

$v$  : the actual flow rate of groundwater ( $\text{m/s}$ )

$t$  : time (s)

$x$  : distance (m)

$\lambda$  : decay constant ( $1/\text{s}$ )

$R_d$  : retardation coefficient

For a continuous source, the analytical solution is obtained as follows:

$$C(x, t) = \begin{cases} A(x, t) & 0 < t \leq t_0 \\ A(x, t) - A(x, t - t_0) \exp(-\alpha \cdot t_0) & t > t_0 \end{cases}$$

#### b. The transport law of radionuclides in the phreatic layer

The two dimensional transport equation for porous medium can be written as

$$D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} - V_x \frac{\partial C}{\partial x} - V_y \frac{\partial C}{\partial y} - \lambda \cdot C \cdot R_d = R_d \frac{\partial C}{\partial t}$$

For a instantaneous release source, the analytical solution is

$$C(x, y, t) = \frac{M}{N_e \cdot R_d \cdot b} X_2(x, t) \cdot Y_2(y, t)$$

The concentration of radionuclides of the continuous source vs time and distance can be obtained by the above equation.

#### c. Dose estimation

The internal exposure by drinking the contaminated water is calculated with the following equation :

$$D^a = C \cdot V^a \cdot g^a \cdot \exp(-\lambda_p)$$

where

$D^a$  : annual effective dose equivalent of individuals in age group  $a$  due to intaking the contaminated water (Sv/a)

$V^a$  : the annual water intake of individuals in age group  $a$  ( $m^3/1$ )

$g^a$  : ingestion dose transformation coefficient in age group  $a$  (Sv/Bq)

$t_p$  : the time interval between getting water from the river and intaking (a)

### 5. 3. 3 Parameters in calculation

#### a. Retardation coefficient

$$R_d = 1 + \frac{\rho}{N_e} \cdot K_d$$

where

$\rho$  : the bulk density of the soil layer medium ( $g/cm^3$ )

$N_e$  : effective porosity of the soil layer medium(%)

$K_d$  : the distribution coefficient of nuclides in the soil layer medium (ml/g)

#### b. Dispersion coefficient

For the homogeneous and isotropic porous medium, the dispersion coefficients are:

$$D_x = \alpha_x \cdot v$$

$$D_y = \alpha_y \cdot v$$

where

$D_x, D_y$  : longitudinal and transverse dispersion coefficients ( $m^2/d$ ) respectively

$\alpha_x, \alpha_y$  : longitudinal and transverse dispersivity (m) respectively

$v$  : actual flow rate of groundwater (m/d)

Table 4 shows the parameters to be used.

### 5. 3. 4 Results

The concentration of radionuclides Sr-90 and Cs-137 in the surface water at the exposed point in Site 1 is shown in Table 5. The dose equivalent of internal exposure resulting from drinking the contaminated water is shown in Table 6. The dose is lower than the prescribed limit, 0.002 mSv/a, of the national standard.

The concentration of radionuclides Sr-90 and Cs-137 on the phreatic water table from Site 2 is shown in Table 7. It can be concluded that there is little impact to the public because of low concentration.

## 6. Conclusions and Suggestions

After the assessment of engineering and radiation impacts, following conclusions are made:

- The natural and social conditions of both Site 1 and Site 2 meet the requirements of national standard GB 9132;
- The radiation impact of waste disposed of at the two sites to the environment is acceptable according to the national standard GB 8703;
- As compared, the design scheme 1 of site 1 is recommended because of its lower investment,

lower disposal cost and shorter distance to the waste source.

Further studies to be made:

- a. Development of safety assessment approaches and the safety assessment of the Northwest Disposal Site;
- b. Establishment of the acceptance criterion of waste for the LLW and ILW disposal sites (especially the limit of TRU contained in wastes);
- c. Performance assessment of the engineered barriers and the overburden layer of the Northwest Disposal Site (including the comparison of the alternative design schemes and their performance assessments);
- d. Development of the inspection technology for waste acceptance in disposal sites;
- e. Establishment of the environmental monitoring plan and the accident emergency measures during the operation and after the closure of the Northwest Disposal Site; and
- f. Development of the computer software for waste management of the Northwest Disposal Site.

Table 1 Site Characteristics

Site Characteristics	Site 1	Site 2
Land slope	1%	0. 6%
Geomorphic stability	Class 2	
Earthquake intensity	<Degree 8(Richter scale)	
Distance to the nearest active fault	1. 5 km	3. 5 km
Distance to the nearest surface water		
perennial river	10 km	2. 5 km
corrasional gully		
Sewer for production sewage	2 km	9 km
Sewer for service sewage	3. 5 km	5 km
Flood risk	no	no
Depth of groundwater	30 m	41 m
Formation of the subsurface	gravel, cobble, clay, sands	gravel, clay, fine sands
Thickness of clay layer (intermittent)	13. 7 m	20. 9 m
Ratio of annual average evaporation to precipitation	58	
Annual average frequency of stationary wind	7%	
Annual average wind speed	5. 8 m/s	
Maximum wind speed	28 m/s	
Useful resources (within 10 km radius)	no	no
Population and residential quarters (within 10 km radius)	no	A residential quarter (36 residents), 3 km away
Utilization of land	no	a little farmland

Table 2 Comparison of the Two Sites

Depth to the groundwater	Site 1 < Site 2
Thickness of clay layer	Site 1 < Site 2
Distance to surface water	Site 1 > Site 2
Distance to reprocessing plant	Site 1 < Site 2

Table 3 The Occupied Areas (O) and the Construction Areas (C) of Each Zone of the Sites ( $m^2$ )

Item	Site 1		Site 2	
	O	C	O	C
Administration and public service zone	864	864	864	864
Operation zone	4,560	1,089	4,560	1,089
Disposal zone	49,104	/	49,104	/
Buffer zone	106,800	/	130,000	/
Total	161,328	1,953	184,528	1,953

Table 4 Parameters in Calculation

Parameters	Site 1	Site 2
Effective porosity	22	20
Bulk density ( $g/cm^3$ )	2.4	1.9
Distribution coefficient of Sr-90 ( $ml/g$ )	0.70	3.3
Distribution coefficient of Cs-137 ( $ml/g$ )	75	500
Retardation coefficient of Sr-90	8.6	32.35
Retardation coefficient of Cs-137	819	4751
Transverse dispersion coefficient ( $m^2/d$ )	0.26	
Longitudinal dispersion coefficient ( $m^2/d$ )	$4.1 \times 10^{-3}$	
Flow rate of ground water ( $m/d$ )		0.16
Thickness of the aquifer (m)		11.3

Table 5 Concentration of Radionuclides in the Surface Water (Site 1) [Bq/m<sup>3</sup>]

Time (a)	Sr-90	Cs-137
50	0	0
100	0	0
200	$9.11 \times 10^{-77}$	0
400	$4.46 \times 10^{-45}$	0
600	$7.99 \times 10^{-9}$	0
730	4.89	0
800	1.39	0
1000	$7.46 \times 10^{-3}$	0

Table 6 The Effective Dose Equivalent (Sv/a) of Various Age Groups

Children	Teenager	Adult
$7.04 \times 10^{-8}$	$3.37 \times 10^{-8}$	$3.53 \times 10^{-8}$

Table 7 The Concentration of the Radionuclides in the Phreatic Water Layer (Site 2) [Bq/m<sup>3</sup>]

Time (a)	Sr-90	Cs-137
50	0	0
100	$1.09 \times 10^{-32}$	0
200	$1.75 \times 10^{-21}$	0
400	$5.22 \times 10^{-13}$	0
600	$3.44 \times 10^{-10}$	0
800	$1.65 \times 10^{-9}$	0
900	$1.36 \times 10^{-9}$	0
1000	$7.29 \times 10^{-10}$	0

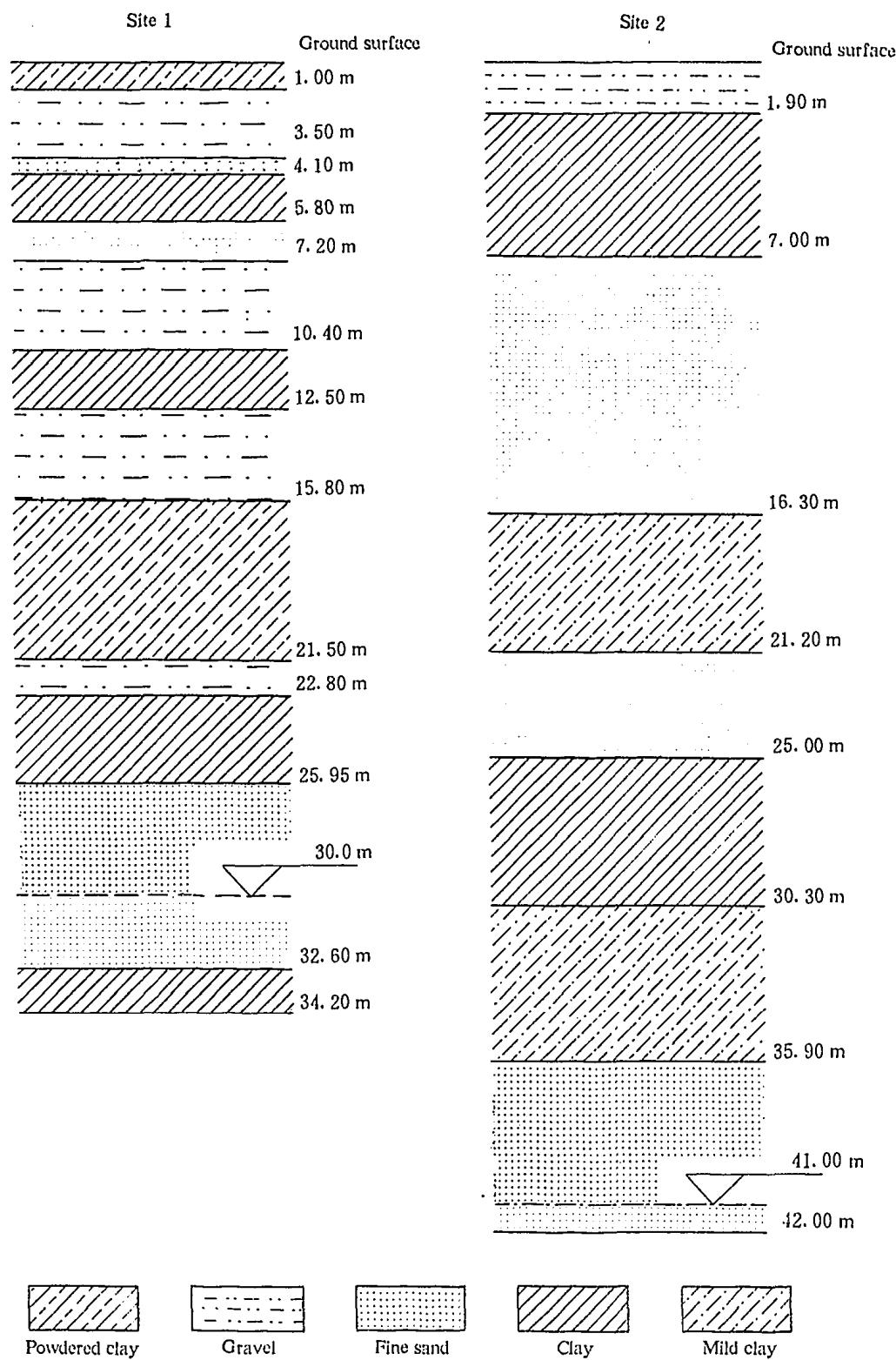


Fig. 1 The Formation of the Subsurface Layers Underneath the Sites

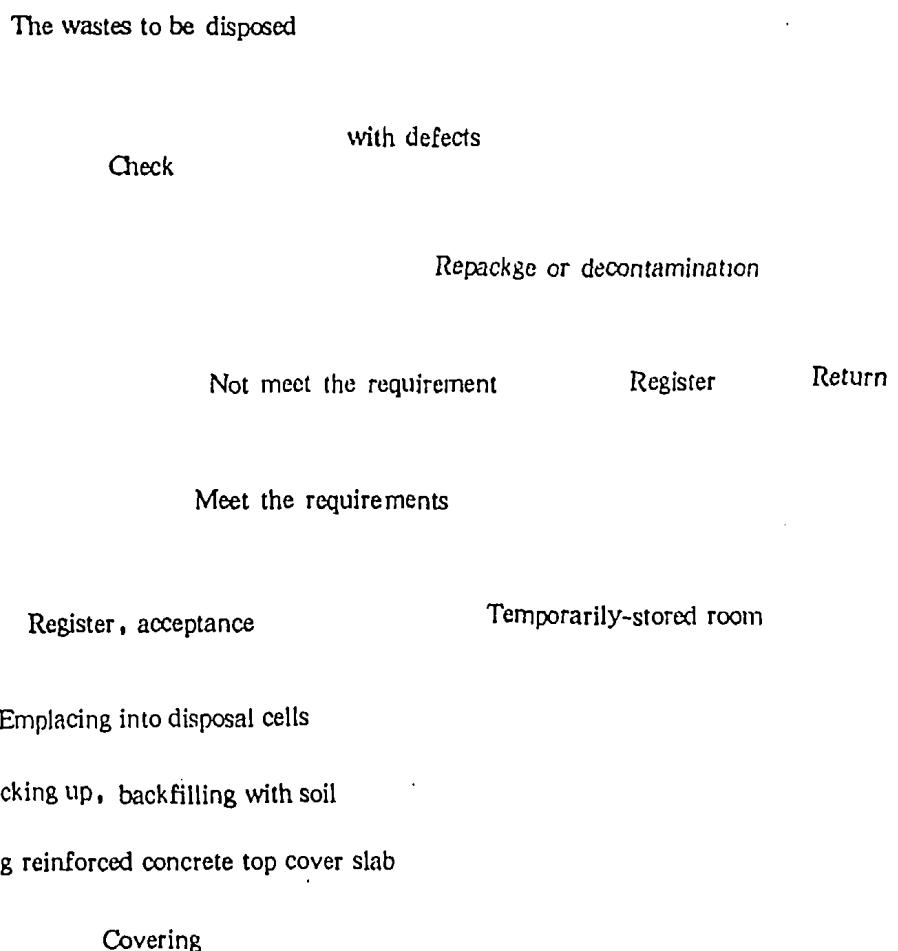


Fig. 2 The Disposal Flow Chart

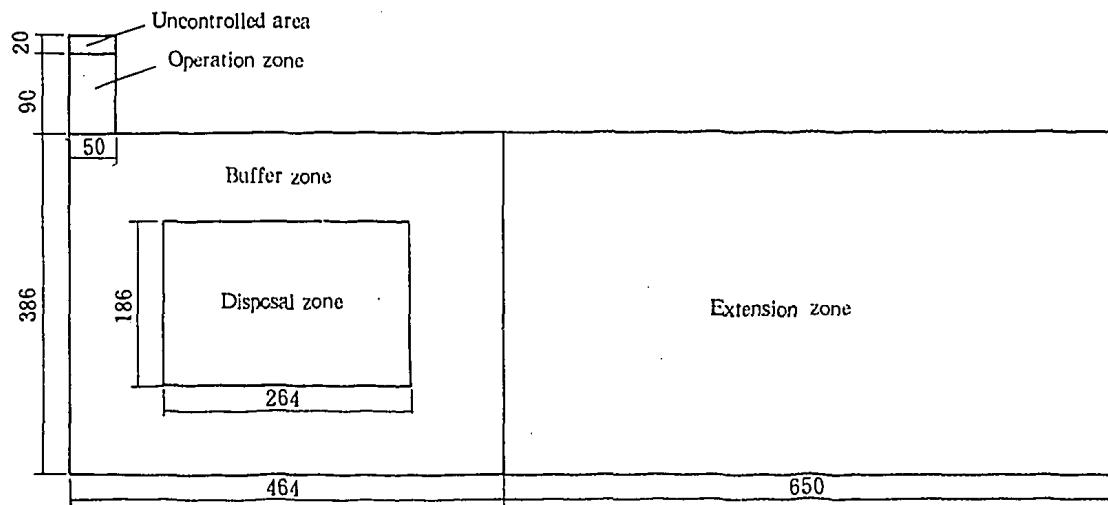


Fig. 3 The Planning Diagram of Site Arrangement (m)

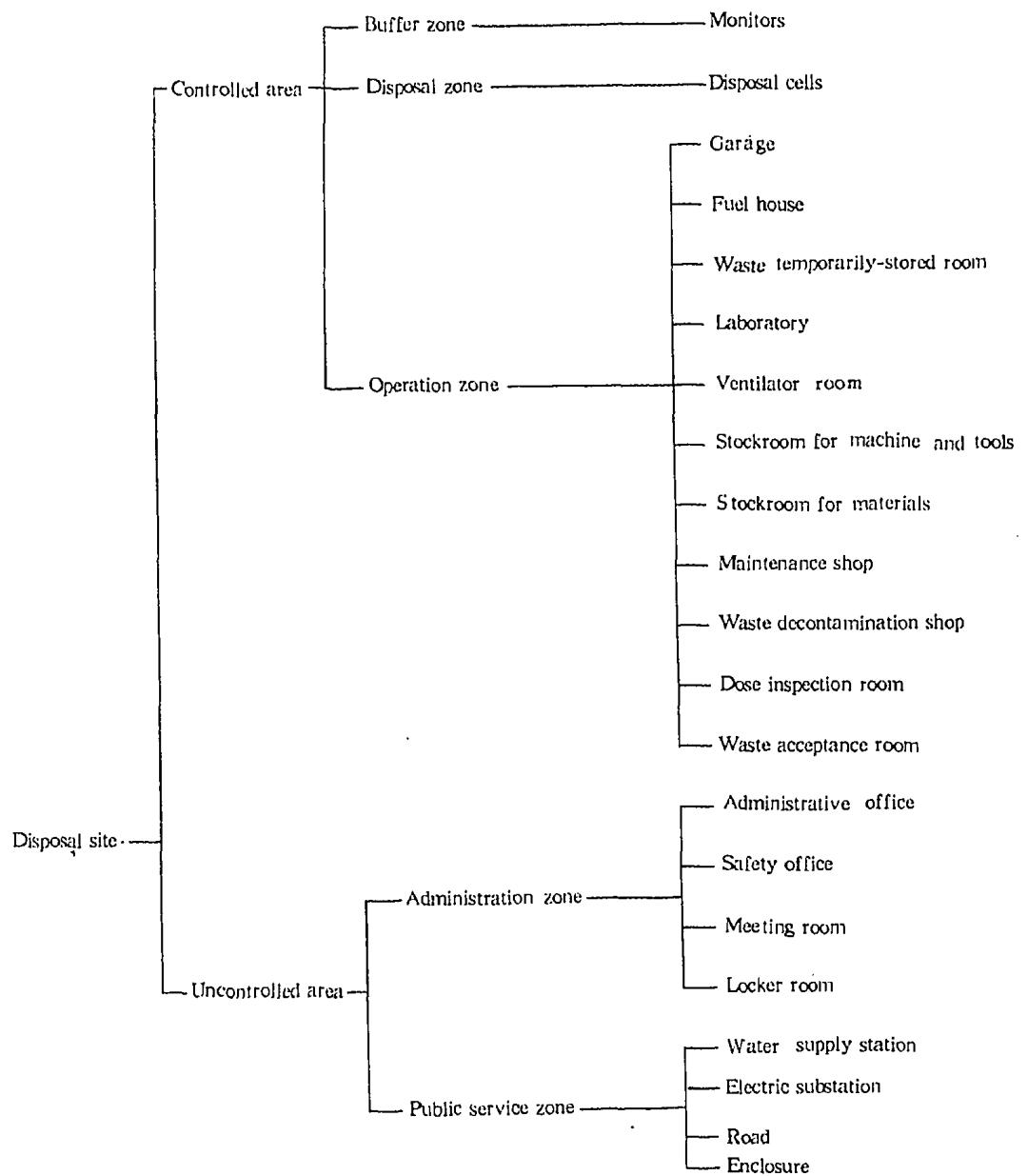


Fig. 4 The Zone Arrangement and the Facilities in the Disposal Site

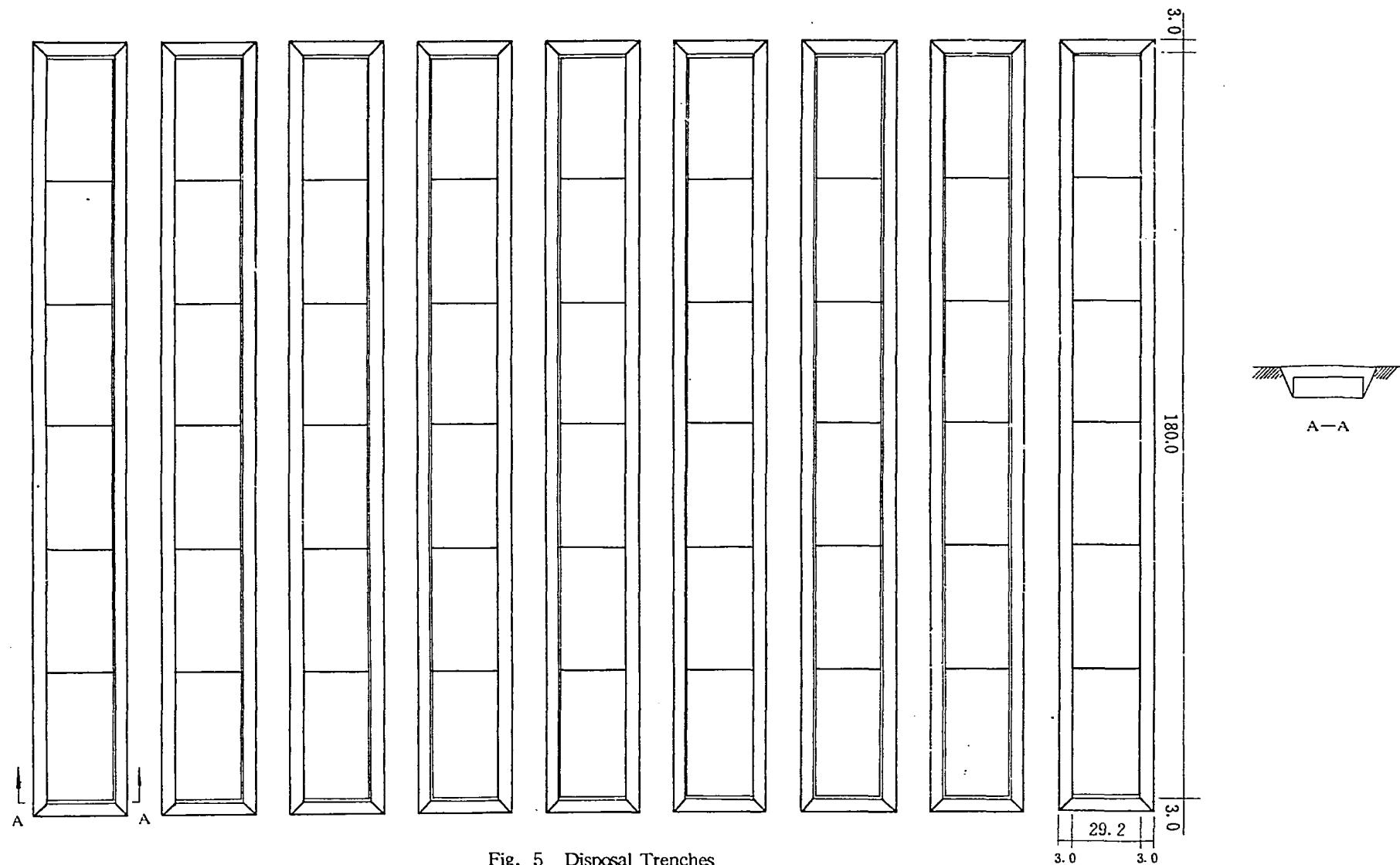
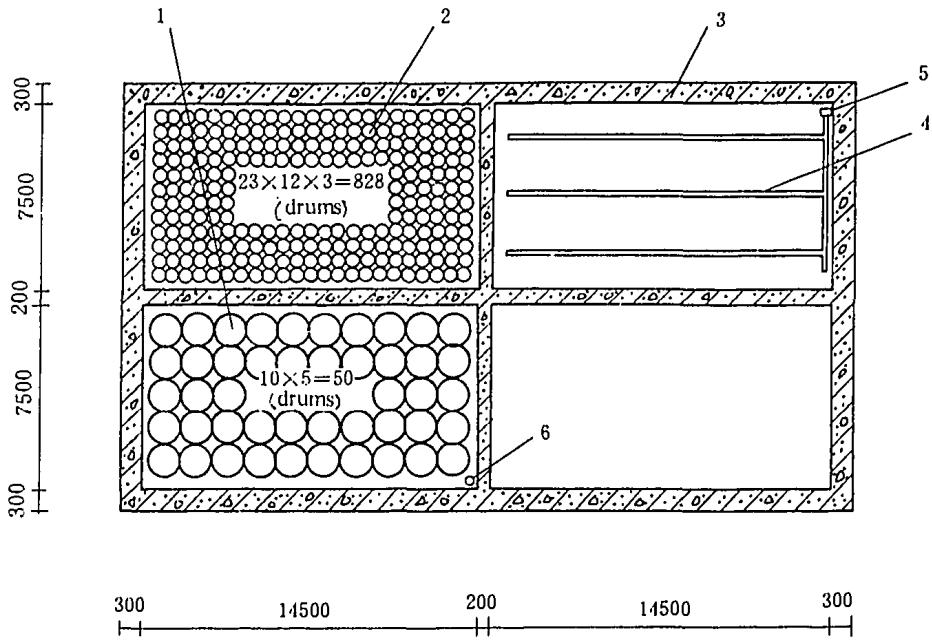
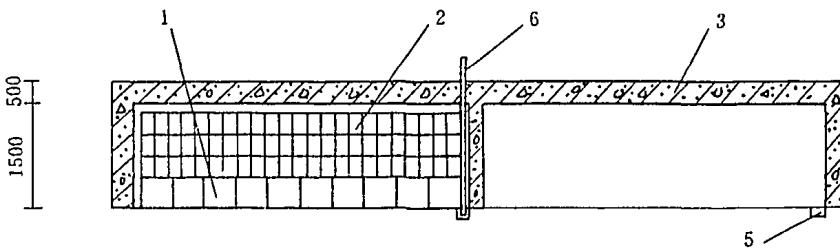


Fig. 5 Disposal Trenches



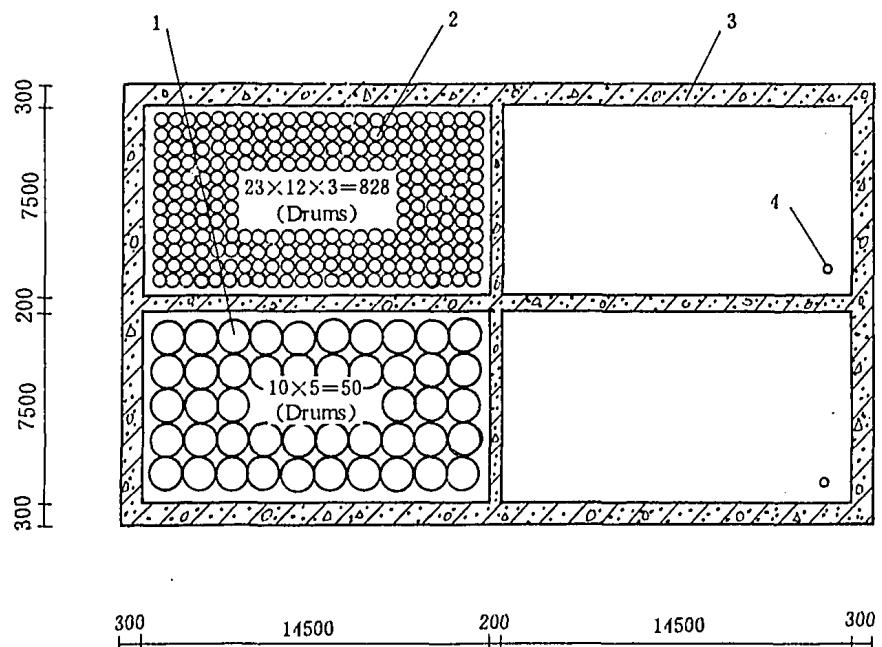
(a) Plane View



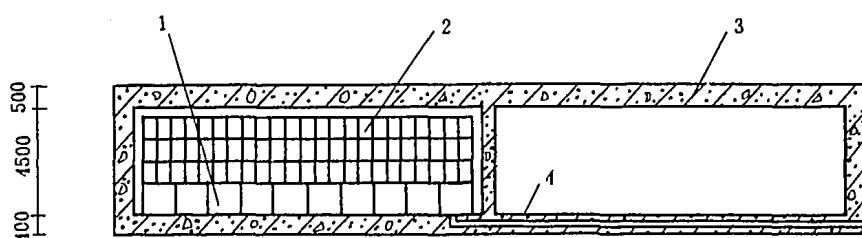
(b) Section View

- |                          |                    |
|--------------------------|--------------------|
| 1. ILW drums             | 2. LLW drums       |
| 3. Engineered barrier    | 4. Drain           |
| 5. Water collecting pool | 6. Monitoring pipe |

Fig. 6 Dimension of Disposal Units (Scheme 1) and Piling of Waste Drums



(a) Plane View



(b) Section View

- |              |                       |
|--------------|-----------------------|
| 1. ILW drums | 3. Engineered barrier |
| 2. LLW drums | 4. Drainage pipe      |

Fig. 7 Dimension of Disposal Units (Scheme 2) and Piling of Waste Drums

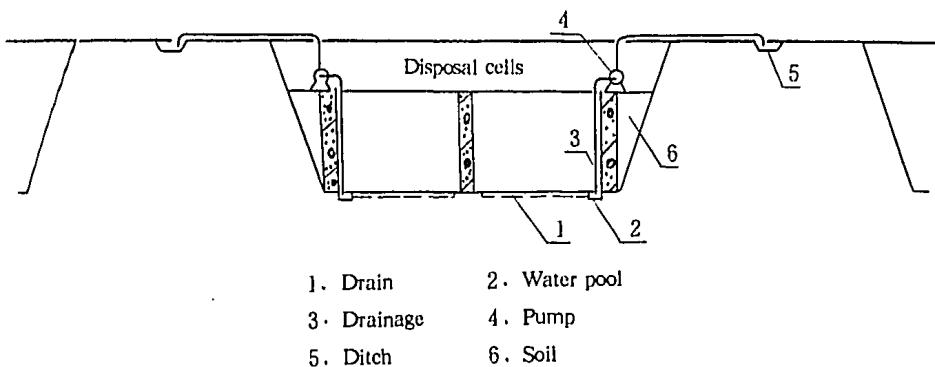


Fig. 8 Drainage System (Scheme 1) of the Disposal Units

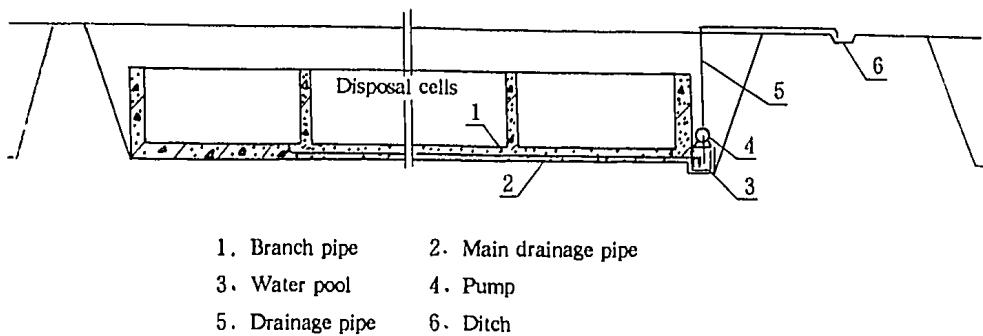


Fig. 9 Drainage System (Scheme 2) of the Disposal Units

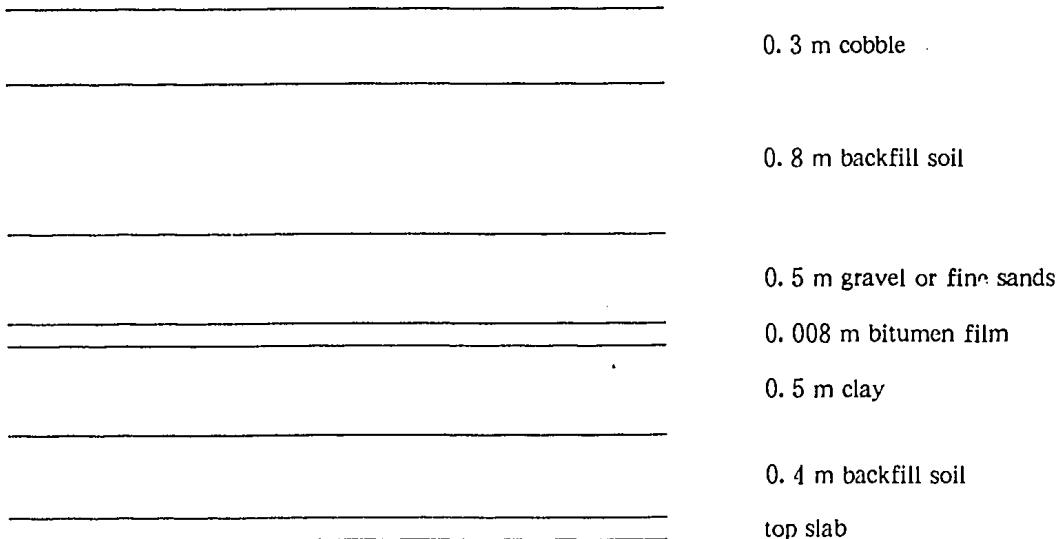


Fig. 10 Section View of the Overburden Layer

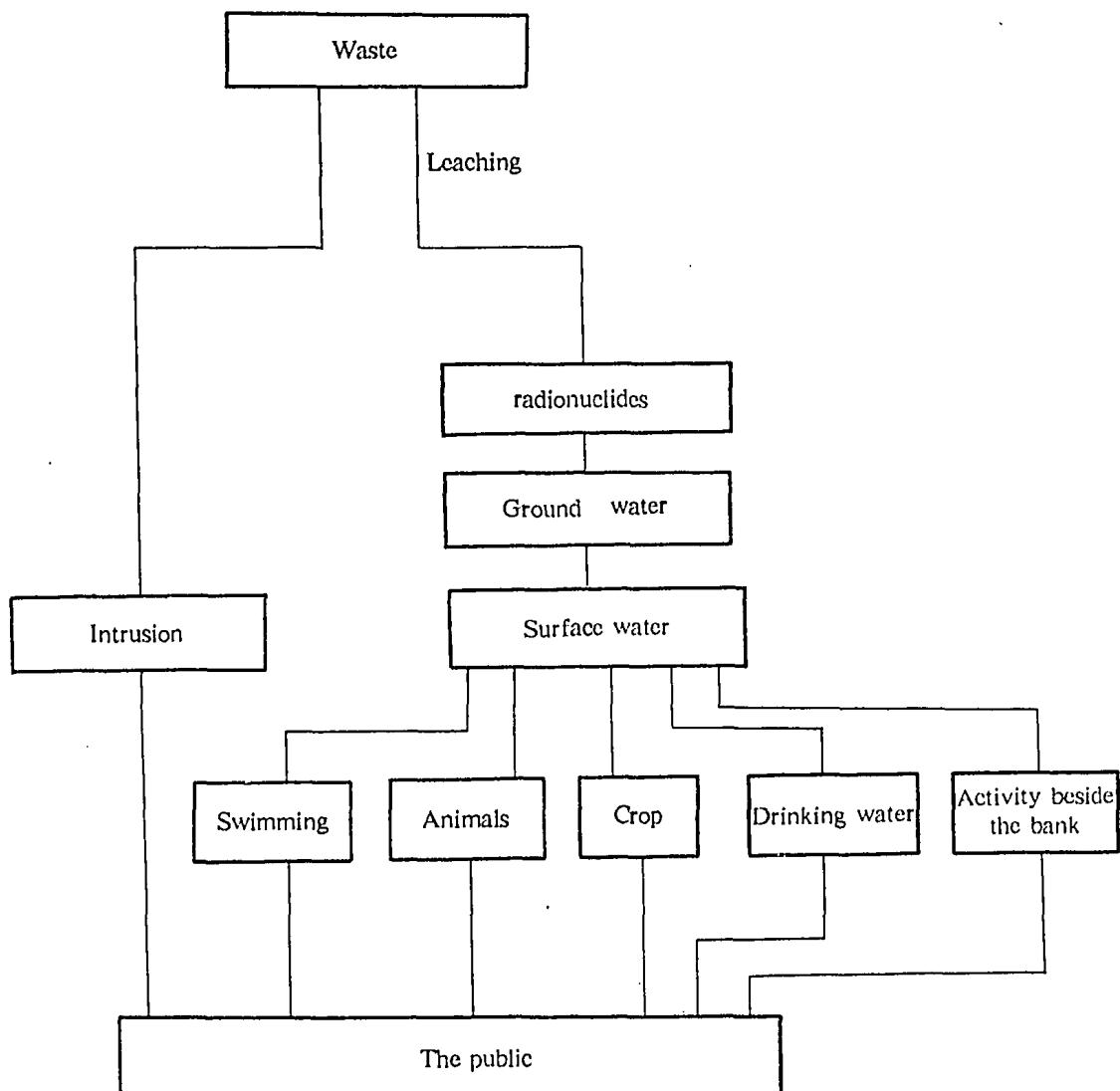


Fig. 11 Potential Pathways of Radiation Impact

### References

- [1] GB 9133 "The Standard for Classification of Radioactive Wastes", The National Environmental Protection Agency, 1988
- [2] "Manual of Radiation Protection", Atomic Energy Public, 1987
- [3] G. H. Firra, et al, "Transport of Radionuclide in Groundwater", 1984
- [4] PAN Ziqiang, "The Assessment of Radiation Environmental Quality on Chinese Nuclear Industry", 1990
- [5] GB 8703, "Regulations for Radiation Protect", The National Environmental Protection Agency, 1988
- [6] GB 9132, "Regulations for Shallow Ground Disposal of Low- and Intermediate-Level Solid Radwastes", The National Environmental Proection Agency, 1988

# **SAFETY ASSESSMENT AND ENVIRONMENTAL MONITORING OF CENTRE DE L'AUBE, FRANCE**

The Monitoring Program and Activities

G. CHEVRIER

International Relations, ANDRA, FRANCE

## **ABSTRACT**

The new low-level waste disposal "Centre de l'Aube", in operation since January 1992, has been characterized in term of environment and activity since the very first stages of the siting process, eight years ago.

The media in which such systematic monitoring is made are divided in two categories; inside the facility, and outside the facility.

Inside the facility, controls are made on surface running water, on infiltrated water collected in the monitoring galleries network, on underground water, on water treatment facility releases, and on storm-basin water.

Also monitored are the sediments and mud from storm-basin and underground galleries, as well as atmospheric dust and radioactivity level at various places.

Outside the facility, controls are made on water from the rivers and from water table, as well as on vegetation and milk.

The actual figures are published by Andra on a three months basis and are widely distributed directly or through the Local Information Commission.

## **1. Main Aims of Near-Surface Repositories**

These aims are set out in basic Safety Rule number I. 2 (RFS I. 2) relating to the design of near-surface repositories.

### **1. 1 First main aim**

The first main aim of a repository is to provide immediate and deferred protection of people and the environment.

Immediate protection is strictly regulated and is applicable to all nuclear facilities.

It was decided to isolate the waste from the environment in order to provide deferred protection for low- and medium-level waste.

This isolation is provided by the actual packaging of the waste and by specially engineered barriers.

The repository and its environment must be monitored for as long as the waste represents a potential hazard.

As a result there are *three periods* in the life of a repository:

First, the operating period during which the waste is placed in storage.

Then the monitoring period which lasts for as long as the distribution of stored materials could present a significant radiological risk. During this phaseperiod only the task of permanent storage in the final state will still be carried out. There will be no more work to carry out, save the possibility of some maintenance or repair work, and the repository will be constantly monitored. Access to the site will be controlled during this period.

Finally, the free access period which begins as soon as the site can be used without any restrictions of a radiological nature for any purpose whatsoever.

The concept of site monitoring prior to the free access period involves the implementation of a programme of checks and maintenance during the entire monitoring period, which explains why a second main aim for the repository was formulated.

### 1. 2 Second main aim

The second main aim of the repository is to allow the site to become harmless at the latest 300 years after the start of the monitoring period.

After it has been rendered harmless, the soil can be used for any purpose whatsoever. The approach adopted to determine the length of the monitoring period consists in studying the radiological impact of the repository in various barrier degradation and soil use scenarios. To retain a wide safety margin, in compliance with RFS I. 2, it is assumed that once harmless, the isolation barriers no longer offer any guarantee of confinement and are no longer protected from human intervention.

Under these conditions, studies of transfers have led to values being set for the maximum admissible quantities for each radionuclide present at the moment when the site becomes harmless, such that the radiological impact remains acceptable.

## 2. Repository Design Bases

Compliance with the main aims results in the following design bases:

a) Isolate the radioactivity during the first two stages in the life of the centre under normal conditions

This in practice involves removing as many of the radionuclides as necessary using outside agents capable of spreading them: water and man.

The risk of the possible dissemination of radioactive substances through water is mitigated by a multiple barrier isolation device comprising modules and engineered structures designed in such a way as to ensure that this device remains effective in all ways for at least 300 years.

To provide protection from any human intrusion, access to the site is controlled until the monitoring period has expired.

b) Limit and/or delay the possible release of radioactivity into the biosphere

To achieve this:

on the one hand, the initial activity of the radionuclides present in the waste modules must be limited,

on the other hand, barriers must be chosen which restrict the migration of radionuclides so that, during the first two periods in the life of the centre, the radiological impact of the repository will remain acceptable under all conditions, and that after the monitoring period the residual activity will be compatible with criteria for rendering the site harmless.

During the operating and monitoring periods, it is the combined properties of the isolation device and the soil and the limitation of initial activity in storage which make it possible to provide an acceptable radiological impact in all plausible accident scenarios.

Once the site has been rendered harmless, even if the engineered structures have not retained 100% efficiency after 300 years, the limitation of activity, the residual properties of the materials from which even deteriorated engineered structures and modules are made, together with the properties of the site, make it possible to keep the radiological risk at an acceptable level.

### 3. Safety Assessment

The safety assessment consists in calculating the individual exposure resulting from a given situation. Generally speaking, the situation is called normal when each barrier is fulfilling its role, and it is called an accident situation when one or more of the barriers loses all or part of its integrity.

Under normal conditions, a very slight fraction of rainwater can leach through the cap (during the monitoring period) or through the soil (during the free access period); this water lixiviates the fraction of waste in the immediate vicinity at that moment; the water then carries the dissolved radionuclides through the basemat, and then through the geosphere to an outlet. The radionuclides are then ingested and inhaled by the critical man.

Accident situations depend to a great extent upon what period in its life the repository has reached:

during the operating period, this might involve:

either handling accidents or fires resulting in workers being exposed to radiation or a spreading of radioactive dust and its subsequent inhalation by members of the public or workers, or accidents jeopardising the integrity of part of the cap, thereby increasing the amount of water entering to lixivate the waste.

during the monitoring period, the most serious type of consequence considered corresponds to a loss of integrity of part of the cap, as a result of a seismic tremor for example,

during the free access period, the situations to be taken into account correspond to intrusions which can be divided into two types:

Situation resulting in an increase, relative to the normal situation, in the amount of contaminated water ingested: creation of a well; in this situation, man short-circuits part of the geosphere by digging directly into the water table, and uses water with a higher concentration of radionuclides by volume.

Situation placing radioactive dust in suspension and causing inhalation, or even exposure to radiation: construction of a motorway, creation of a residential area, etc. The various possible hazardous elements are given in Table 1. Only those events which have a probability of occurring of greater than  $10^{-5}$  per year have been included.

To be in a position to give a quantitative assessment of the radiological impact of the repository for the various scenarios chosen, the processes by which the radionuclides are transferred to man have to be analysed.

Three processes can be distinguished:

- transfer by water,
- transfer by air, and
- external exposure.

### 3. 1 Analysis of transfer by water:

#### 3. 1. 1 Activity carried every year by infiltration water

The infiltration water can carry over, by a process of lixiviation, a certain amount of radionuclides. To characterise this carryover a magnitude known as the *lixiviated activity fraction* is used every year. It is defined, for a given radionuclide as the ratio of activity lixiviated every year to the residual activity of the module at the time of lixiviation. The amount of activity carried over every year is calculated for a given radionuclide as the product of the following factors:

Cumulative fraction of modules with a deteriorated overpack, the contents of which might be exposed to the action of water for the given year; it depends on the ageing characteristics of the module overpack (concrete or metal container).

The annual lixiviated activity fraction: this is dependent upon the amounts of water which have infiltrated through the cap, and upon the nature of the materials surrounding the waste. Its value differs depending upon what stage of life the repository is in and on the way in which the waste is stored and packaged.

The residual disposal activity for the year in question.

#### 3. 1. 2 Modelling the transfer of radionuclides into the soil

The method adopted consists firstly in characterising the flow of water into the soil by a velocity field  $V_p$ , then in studying the migration of radionuclides into the soil in light of the retention and spreading phenomena which characterise it.

The flow is supposed to be on-going and two-dimensional; it can be characterised at any point by the permeability of the aquifer,  $k$ , the hydraulic load in the table  $h$  (piezometric levels) and the porosity  $w_c$ . The real flow velocity is written in the form:

$$V_p = -\frac{k}{w_c} \text{ grad } h$$

Owing to physical and chemical interactions between the natural environment and the radionuclides, the average velocity of the radionuclides is generally much lower than the real velocity of the water (except for tritium).

The real velocity is obtained by dividing the velocity of the water by a delay co-efficient which is a function of the ratio of distribution between the water and the soil. This ratio is specific to each radionuclide. It is a function of the properties of the soil.

The transfer calculations are carried out using a numerical code which supplies:

- the quantities of activity released annually by the repository, and
- the quantities of activity transferred via the outlet every year in the light of the soil

properties.

### **3. 1. 3 Modelling the transfer of radioactivity into the biosphere**

In order to simulate the variety of routes and mechanisms which are involved in the dispersal of radionuclides into the environment, the following transfer paths were adopted:

- release and dilution of radioactivity into fresh water,
- migration of radionuclides into the soil after irrigation or flood,
- transfer of radionuclides into the environment to non-aquatic plants,
- transfer into animal products, and
- ingestion or inhalation by man of products possibly containing radionuclides.

A calculation code, which adopts the conventional equations for the transfer of radioactivity into the biosphere, was used. The values of those parameters which depend specifically on radionuclides come from an artificial CEA library.

### **3. 2 Analyses of transfer by air**

Dispersion or the placing into suspension of waste can occur:

- during the operating period in the event of a module being dropped or set alight. The activity inhaled depends on the distance from the source and on the atmospheric dispersal conditions at the scene of the accident, and
- during the free access period, the risk of inhaling radioactive dust can affect workers carrying out earth moving work, civil construction work, members of the public living on-site or nearby and likely to inhale dust placed in suspension by the wind.

### **3. 3 External exposure**

During the free access period, the site is considered to no longer offer any protection from human intrusion. It is assumed that the waste is mixed with degraded material from the engineered disposal structures and from the original soil.

External exposure is in essence a result of gamma radiation, emitted by the contaminated soil, which, in view of its thickness, can be compared with a semi-infinite environment.

## **4. Radiological Monitoring**

Repositories are subjected to radiological monitoring to protect man and his environment from the risks caused by the activities of packaging and storing radioactive waste.

The aims of site and environment monitoring are:

- to comply with the requirements imposed on the nuclear operator by the regulations and provisions issued by the safety authorities in the light of the waste characteristics and potential transfer paths for radioelements from the repository to man, and
- to detect, analyse, and correct any problem which could arise, equally well in radiological fields as in non-radiological fields.

The radiological monitoring of a repository and its environment are covered by a regulatory radiological monitoring plan submitted to the authorities for approval.

The environments affected by the checkings are:

On-site:

runoff water, infiltration water passing through the engineered disposal structures and collected by the buried gravity separation pipes, groundwater, and water from the biological purification plant and from the storm basin,  
sediments and muds from the storm basins and the buried gravity separation pipes,  
atmospheric deposits, and  
radiation at the outer enclosure.

**Off-site:**

water from streams and groundwater,  
vegetation, and  
milk.

Before the Centre de l'Aube repository was brought into service, a radiological reference state (point zero) was established.

This was intended to evaluate, before the repository came into service, the relative shares of naturally and artificially occurring radiation in the main components of the earth and water systems. For the Centre de l'Aube repository, the surveys, samples, and measurements were taken in two stages:

- a preliminary point survey intended to provide an initial evaluation of the levels of radioactivity for a few samples as well as the ambient gamma irradiation values (November 1985), and
- a radioecological study over a period of time longer than the previous one in order to take in the seasonal and plant cycles (from June 1986 to June 1987).

More than 1500 measurements were taken.

### **Conclusions**

In the beginning of January 1992 the Centre de l'Aube repository started operation. A certain number of technical solutions were added during the design stage for increased security: automatic measuring devices were developed to limit the exposure of workers to radiation, buried gravity separation pipes were included to enable the collection of water which could have infiltrated into the engineered structures, modules and engineered structures with confinement properties good for over 300 years, the site having some radionuclide retention properties.

A method taking into account the experience obtained during operation of the Centre de la Manche repository has been gradually developed for the safety assessment. Analyses carried out on behalf of the Centre de la Manche and the Centre de l'Aube, have shown that the radiological impact on the populations is negligible for the normal evolution scenario and also for the so-called accident scenarios.

Table 1 List of hazardous events

Source of the event	Operating	Stage monitoring	Rendering harmless
Human	<ul style="list-style-type: none"> <li>-Handling incident,</li> <li>-Fire (waste, facility),</li> <li>-Manufacturing defect</li> <li>(modules, basements, structural concrete, cap, water collection pipes),</li> <li>-Plane crash</li> </ul>	<ul style="list-style-type: none"> <li>-Manufacturing defect (see previous column),</li> <li>-Plane crash</li> </ul>	<ul style="list-style-type: none"> <li>-Intrusion: Site</li> <li>Residential area</li> <li>Children's play area</li> <li>Wells</li> <li>-Plane crash</li> </ul>
Natural	<ul style="list-style-type: none"> <li>-Earth tremor,</li> <li>-Flood,</li> <li>-Intrusion by animal,</li> <li>-Intrusion by plants,</li> <li>-Severe weather conditions,</li> <li>-Meteorite fall</li> </ul>	<ul style="list-style-type: none"> <li>-Earth tremor</li> <li>-Flood</li> <li>-Intrusion by animal</li> <li>-Intrusion by plants</li> <li>-Severe weather conditions,</li> <li>-Meteorite fall</li> </ul>	<ul style="list-style-type: none"> <li>-Earth tremor</li> <li>-Flood</li> <li>-Intrusion by animal</li> <li>-Intrusion by plants</li> <li>-Severe weather conditions,</li> <li>-Meteorite fall</li> </ul>

# **ENVIRONMENTAL POLICY ON RADWASTE MANAGEMENT AND DISPOSAL IN CHINA**

**ZHAO Yamin**

National Environmental Protection Agency, China

## **ABSTRACT**

This paper introduces the environmental policy on radwaste management and disposal.

In order to prevent different kinds of radwaste from polluting environment, ensure public health, and simultaneously promote the development of nuclear energy and nuclear technology, a set of environmental policies on radwaste management and disposal has been established. The major policy are as follows:

- Solidifying the temporarily-stored radioactive liquid waste as early as possible
- Limiting the temporarily-stored time for intermediate-and low-level solidified radwaste, and solid radwaste.
- Constructing regional disposal repository for Low and Intermediate level radwaste (L/ILW)
- The radwaste and spent radiation sources arising from nuclear technology application shall be sent to the provincial waste repositories that are named City Radwaste Repository.
- The radwaste coming from the development and application of intergrown radioactive mineral resources should be stored in the dams which have to be provided.

## **1. Introduction**

The development of nuclear power and nuclear technology brings a great of benefit to the mankind, but, at the same time, it produces the radwastes. Nowadays, all countries pay great attention to the management of radwaste. "The Agenda of the 21st Century" mapped out on the World Environment and Development Conference in Brazil in 1992, has a special chapter discussing the environmental management of radwaste. This shows that radwaste has became the important subject of environmental protection. China's nuclear industry and nuclear technology application has a history of dozens of years. The environmental management of radwaste has been thought highly of in a very early date. In 1982, the National Environmental Protection Agency (NEPA) was authorized to take the responsibility for managing the radwaste of the whole country. This paper is intended to introduce simply the management and disposal policy on the radwaste of our country.

## **2. China's Environmental Protection Authorities Responsible for the Management of Radwaste**

"The Environment Protection Law of PRC" (trial version) (1979) stipulated in its No. 16 Article the key elements of environmental pollution to be prevented and treated;

"Actively prevent and treat the pollution and harm of harmful substances such as; waste gas, liquid waste, waste residue, dust, garbage, radiation substance etc, noise, vibration and foul smell produced from industrial and mining enterprises and cities to the environment."

In this Article, the radioactive substance is stipulated together with other harmful substances as the contents of environmental protection.

Soon afterward, in the 26 Article of the Law, it also defines the organization of the environmental protection and its function:

"The State Council establishes the environmental protection organization, whose main duties are:

- (1) Implementing and supervising the execution of State's general and specific policies, law, and decree on environmental protection;
- (2) Mapping out, jointly with other departments concerned, environmental protection regulations, stipulations, standards, and economic and technical policies;
- (3) Formulating, jointly with other departments concerned, long-term program and annual plan; and supervising and inspecting their execution;
- (4) Carrying out environmental monitor under unified organization; investigating and mastering status and developing tendency of country's environment; and putting forward the improvement measures;
- (5) Organizing and coordinating, jointly with other departments concerned, environmental science research and environmental education cause. Actively popularizing domestic and foreign advanced experience and technique of environmental protection;
- (6) Giving guidance to the subordinate departments of the State Council, provinces, autonomous regions, and municipalities on the work of environmental protection; and
- (7) Organizing and coordinating international cooperation and exchange of environmental protection."

Combining these articles, it shows that the Environment Protection Law has authorized the environmental protection authorities to assume the responsibility for preventing and treating radioactive contamination.

According to the Environment Protection Law (trial version), the NEPA was established in 1982, which took charge of executing the tasks that were authorized by the Law. On October 19 of the same year, the then Secretary-General suggested the proposal as follows:

"The Environmental Protection agency will take charge of the centralized management of whole country's radwaste. It's main tasks are; unified planning, organizing coordination, and supervising and inspecting the activities of environmental protection. The Ministry of Nuclear Industry (the predecessor of China National Nuclear Corporation), the Ministry of Metallurgical Industry, the Ministry of Public Health, and departments and regions engaging in radioactive activities should take charge of the arrangement of radwaste disposal and storage of their own system and regions."

On October 21 of the same year, the State Council approved the above proposal. The NEPA is responsible for the centralized management of country's radwaste.

### 3. Sources of Radwaste and Their Classification in Light of Management

In China, the radwaste mainly comes from the China National Nuclear Corporation (CNNC), the Ministry of Metallurgical Industry (MMI) and the Ministry of Public Health (MPH) (see Fig. 1).

The CNNC owns uranium mines, refineries, hydrometallurgical mills, enrichment plants, fuel element plants, various kinds of reactors and reprocessing plants. Now nuclear power plants are also started to be built. In the past 30 years, the CNNC produced numerous radwaste, which is referred as the nuclear facility radwaste (NFRW).

The MMI has non-ferrous metal ore deposit and rare earth ore deposits (China is rich in rare earth). Some non-ferrous metals and rare earth elements intergrow with uranium and thorium during mineralization. In mining those ore deposits, the underground associated uranium will be brought to the surface, sometimes the radioactivity content of tailings may reach the radwaste level. In regard to such radwaste, the management should be enhanced too. This kind of radwaste is referred as the intergrown ore radwaste (10 RW).

The MHP uses nuclear technology for diagnosis and therapy. Besides, the nuclear technology is also used in agriculture, industry, scientific research and education. These activities may also produce radwastes named nuclear technology application radwaste (NTARW).

#### 4. Environmental Policy on Radwaste Management and Disposal

Authorized by the State Council, the NEPA is responsible for the centralized management of whole country's radwaste. In order to have an efficient management, we pursue the criss-cross type managing approach, of which the vertical aspect refers to the management of every stage from radwaste production to treatment, storage, transportation, until to final disposal, while the horizontal aspect refers to as such management as formulating the standard, reviewing and approving the environmental impact report, carrying out on-site supervision and monitor etc. Fig. 2 shows the major points of horizontal management.

The purpose of criss-cross management system is to effectively protect the environment, and effectively prevent radwaste from entering the living environment of mankind. The corresponding environmental policy of different kind of radwastes has been worked out.

##### 4. 1 Management and disposal policy for the L/ILW of nuclear facilities

The disposal policy for the L/ILW of nuclear facilities has experienced a course of development. In the early stage, the radwaste was placed on the spot near the facility. In recent years, after studying the transportation cost and the risk of radwaste, the regional disposal is believed to be feasible. Now, the regional disposal policy of L/ILW for the nuclear facilities has been approved by the Chinese government. The main points are as follows:

- Solidify the temporarily-stored radioactive liquid waste as early as possible.
- Limit the temporary storage time of intermediate and low-level radioactive solidified waste and solid waste. The L/ILW of nuclear power plant is defined as temporarily storage for 5 years.
- Construct the regional disposal repository of L/ILW to dispose the waste to be produced in this area and its adjacent areas.
- The CNNC takes the responsibility of siting, constructing and operating of the regional disposal repository of L/ILW.
- The NEPA is responsible for reviewing and approving the environmental impact reports of disposal repository; formulating and promulgating relevant standards and regulations and guidelines; the administrative competent authorities of environmental protection of provinces, autonomous regions and municipalities are responsible for supervising the environ-

mental protection activities of disposal sites.

-Affirmation of fund raising channels.

The long-term loans shall be arranged by the relevant departments of the state; and part of the capital construction cost of nuclear power plant shall be allocated as the initial fund, which is used mainly in the pre-stage work, construction and initial operation.

The disposal repository, as constructed, provides service on the basis of compensatory approach. The income collected herefrom will be used to pay off the loans, and maintain the operation.

#### 4. 2 Environmental policy on high-level radwaste disposal

The high-level radwaste includes spent fuel, to which the policy is temporarily stored with care, while those liquid radwaste shall be solidified. The deep geological disposal are adopted at dozens of years later.

#### 4. 3 Management and disposal policy for NTARW

As early as 1987, China had published the management criteria for NTARW whose major points are as follows:

- All users of radioisotopes and nuclear technology including those in industry, agriculture, medicine, scientific research and education departments where radwaste and spent radiation sources are produced shall adhere to this criteria.
- The environmental protection authorities of each province, autonomous region, and municipality, special organization shall be set up and staffed with professionals to take charge of centralized supervision, management, and environment monitor of the radwaste from nuclear technology application.
- Any contaminant containing artificial radionuclides with specific activity greater than  $2 \times 10^4$  Bq/Kg, or containing natural radionuclides with specific activity greater than  $7.4 \times 10^4$  Bq/kg, shall be treated as radwaste.
- The radwaste from nuclear technology application can be classified as three categories in terms of their half-lives:

Short-life radwaste  $T_{1/2} \leqslant 60$  days

Medium half-life radwaste  $60 \text{ days} < T_{1/2} \leqslant 5.3 \text{ years}$

Long half-life radwaste  $T_{1/2} > 5.3 \text{ years}$

- Any user producing radwaste shall take all necessary measures to minimize waste amount or waste volume. It is not allowed to dispose radwaste and spent radiation sources in the environment by user itself. They shall be collected and disposed in a centralized way by the management authorities for radwaste from nuclear technology application.
- The radwaste shall be well-packed. The surface dose rate of each bag shall not exceed 0.1 mSv/h, the volume is controlled under 30 l, and the weight, under 20 kg. The external surface contamination of the package shall be controlled to the following levels respectively:  
 $\alpha < 0.04 \text{ Bq/cm}^2$        $\beta < 0.4 \text{ Bq/cm}^2$

The external surface dose rate of the special vehicle for transportation shall be lower than 0.2 mSv/h, and that in the driver's cab shall be lower than 0.025 mSv/h.

- The packed radwaste, when entering the waste repository shall be inspected and accepted individually. The registration cards shall be well filled, and the time of retaining the cards shall not be less than the time that the radwaste becomes harmless.
- The packed radwaste in the waste repository shall be classified and stored according to the stip-

ulation. Those radwaste and spent radiation sources which can not decay to  $\leq 2 \times 10^4$  Bq/kg within the safe storage period can only be stored temporarily in the waste repository, and they shall be ensured to be withdrawn for transferring to the final disposal repository.

-During the storage period any radwaste decayed to below  $2 \times 10^4$  Bq/kg can be buried as common garbage in a simply constructed pit within the repository area, after being reported to and approved by the environmental protection authorities of the province or autonomous region or municipality concerned.

-The environment inside and around the repository area shall be monitored periodically.

#### 4. 4 Policy on intergrown ore radwaste

- (1)The large amount of radwaste formed during mining shall be stored in the tailings dam.
- (2)The small amount of radwaste formed during the processing of rare earth shall be stored temporarily in the waste repository for NTARW, and later shall be sent to the nearby tailings dam for NFRW or for IORW.

### 5. Implementation of Environmental Policy on Radwaste Management and Disposal

According to the Environment Protection Law, the authorities of environmental protection are responsible for the centralized supervision and management of radwaste, while the competent authorities of the users that produce the radwaste are responsible for the management of their own radwaste. On the basis of this principle, in the execution of environmental policy on radwaste management and disposal, two aspects of initiative have to be brought into play: one is the role of environmental protection authorities; and the other is that of the users that produce the radwaste, and their competent authorities.

Besides, in regard to the high-level radwaste, especially the spent fuel, the issues of nuclear safety and substantial guarding are involved with. Also, the spent radiation sources arising from nuclear technology application has the problem of security and guarding. Referring to such problems, the environmental protection authorities have to cooperate closely with the National Nuclear Safety Administration (NNSA) and the Ministry of Public Security (MPS) to arrange the work concerned well. Fig. 3 shows the relationship between the NEPA, the NNSA and the MPS on the management of radwaste.

For effectively implementing the environmental policy, it is intended that different management is to be used for different kind of waste. Figs. 4, 5, 6 and 7 give respectively the management flow charts of L/ILW of nuclear facilities; high-level radwaste of nuclear facilities; radwaste from nuclear technology application and radwaste from intergrown ore. At present, the management of radwaste have run according to these procedures. Practice proves that China's environmental policy is effective and feasible.

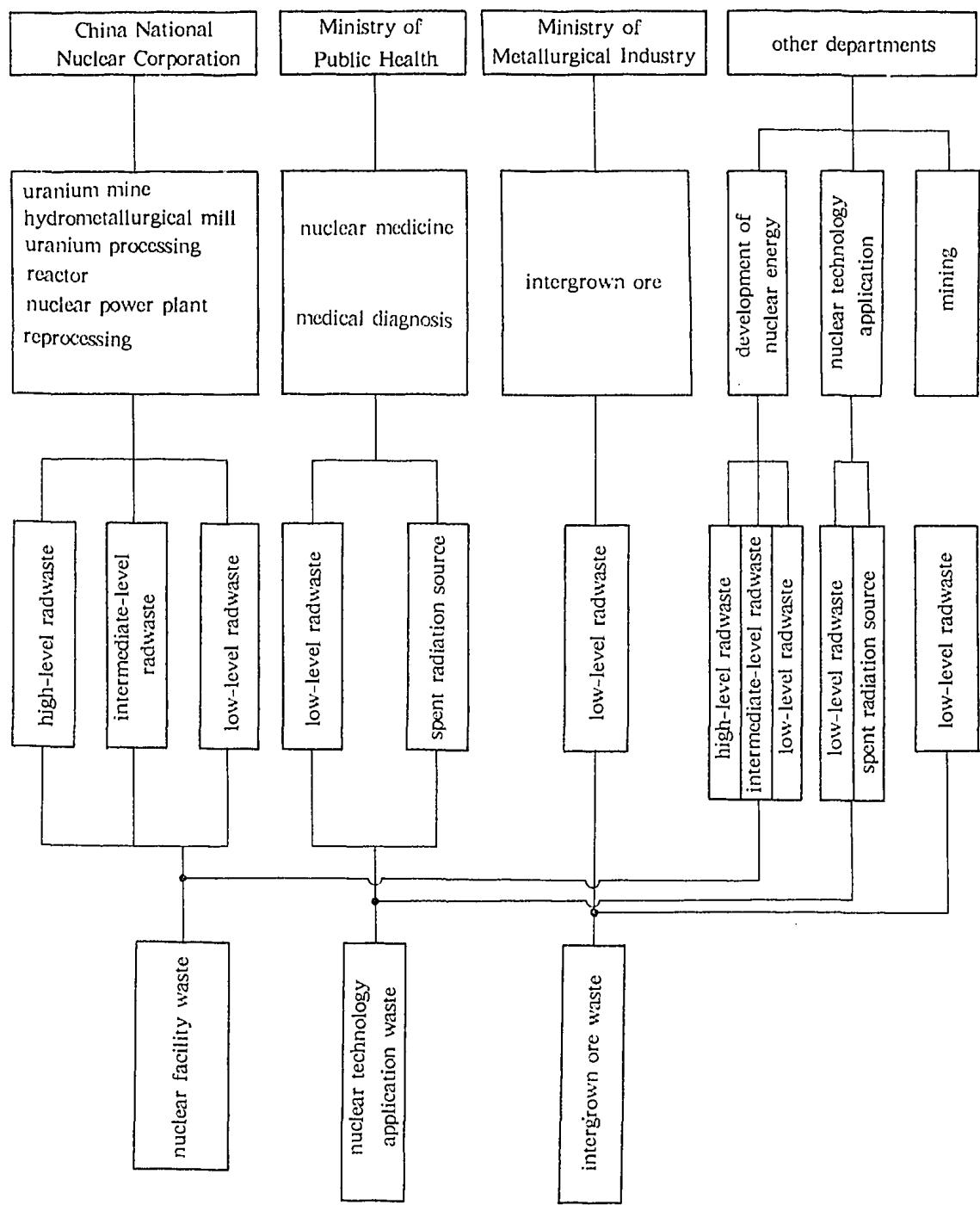


Fig. 1 Production channel of radwaste

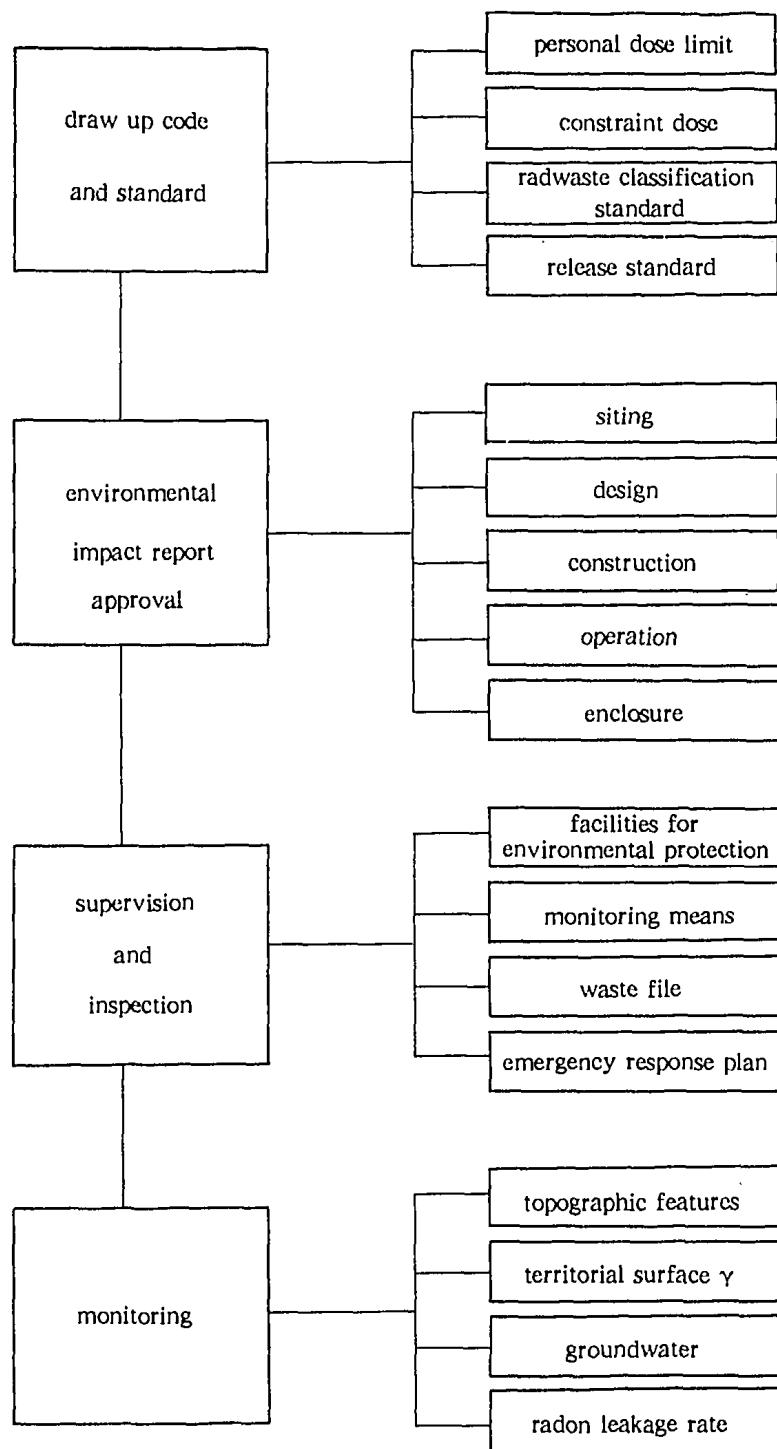


Fig. 2 Main points of environmental protection management of radwaste disposal

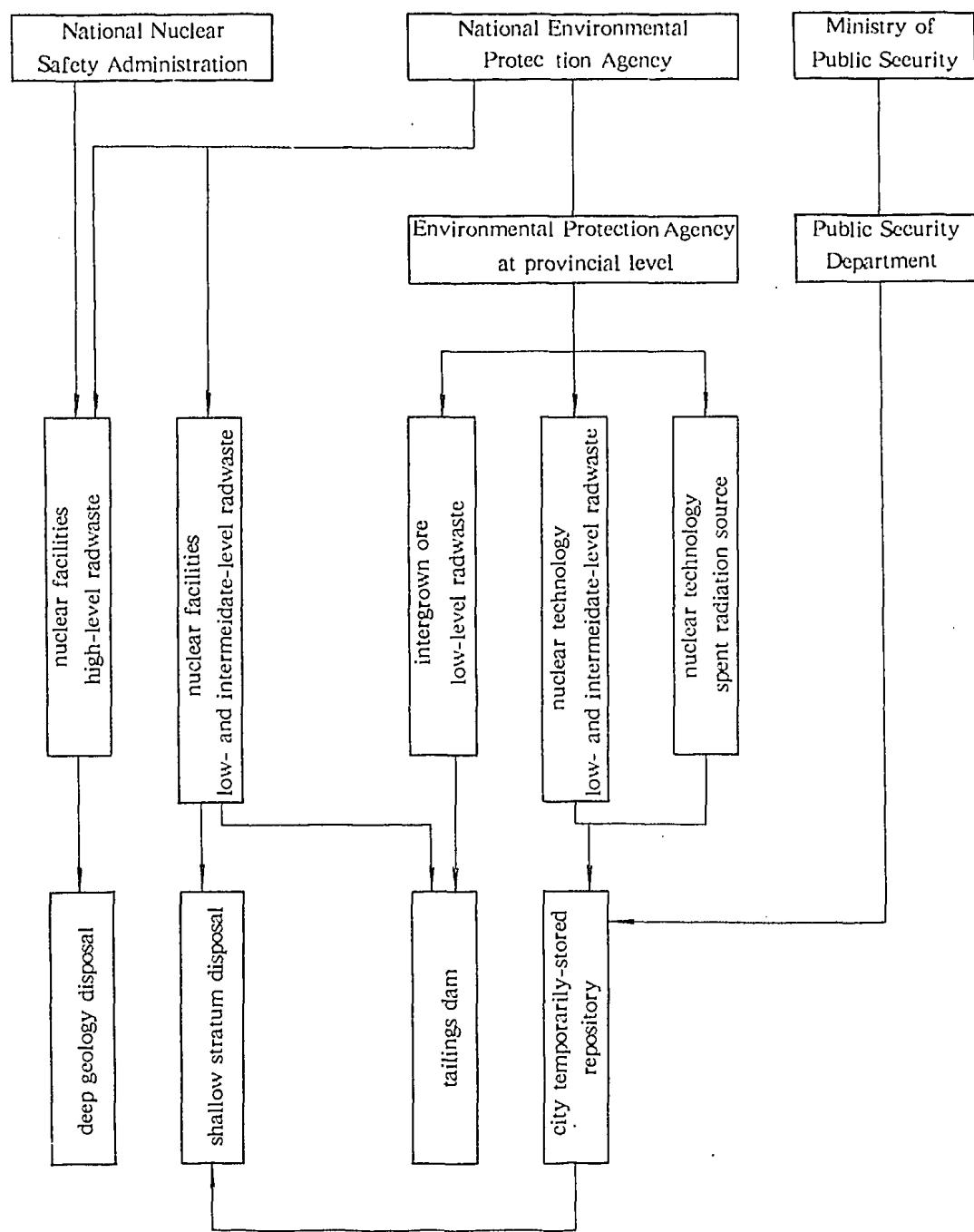


Fig. 3 Relationship chart of management for waste disposal

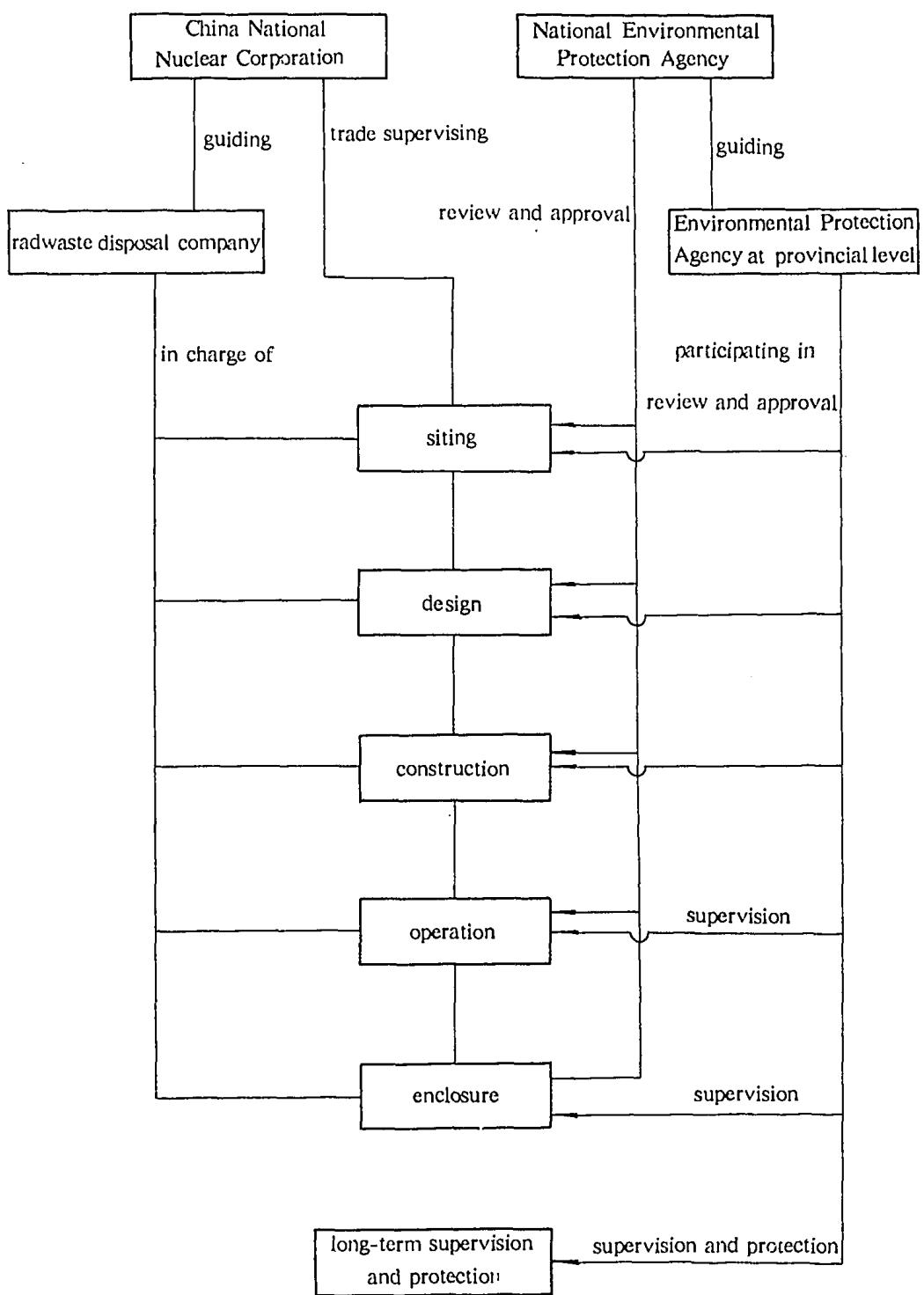


Fig. 4 Management flow chart for intermediate-and low-level radwaste disposal from nuclear facilities

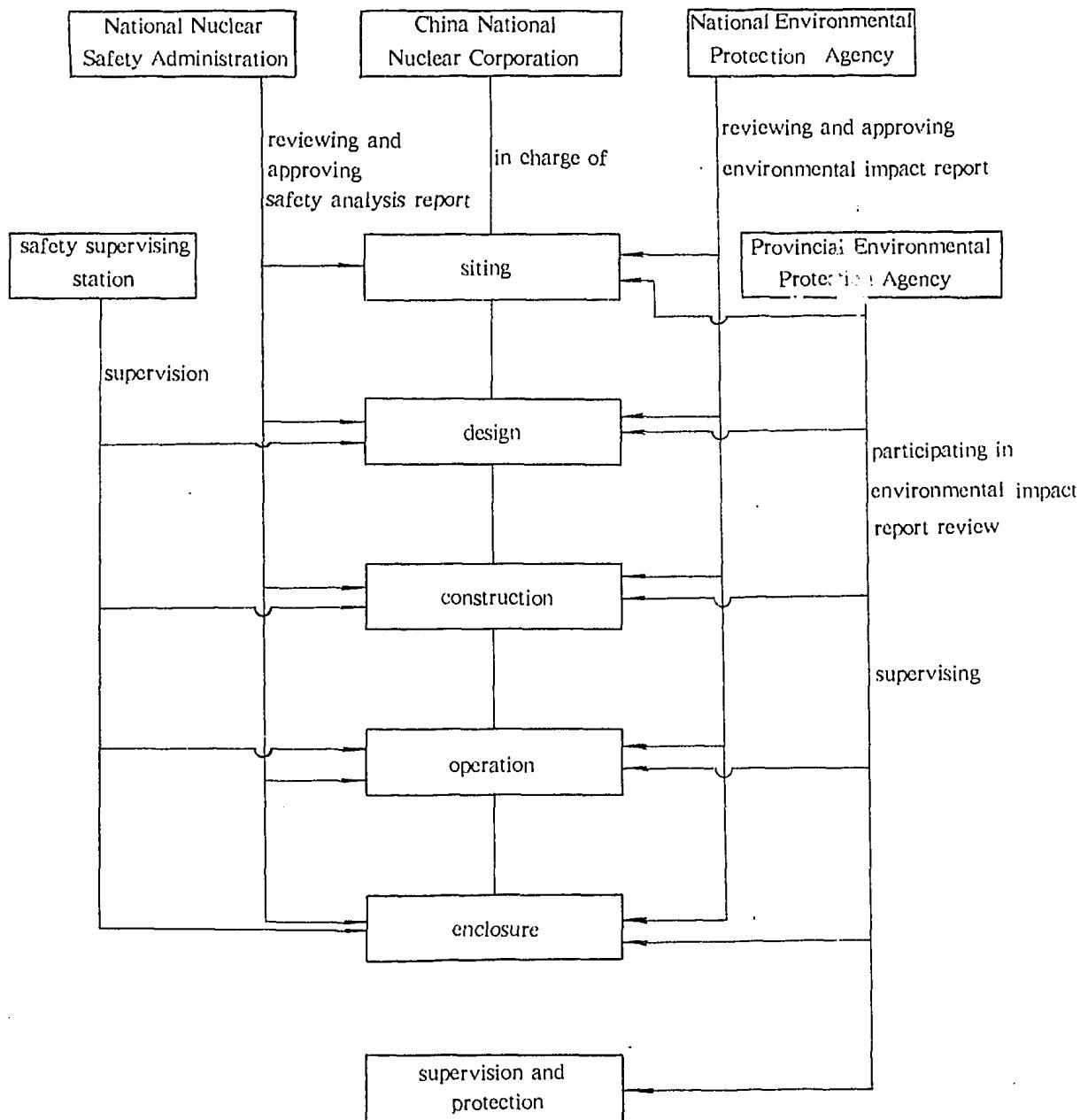


Fig. 5 Management and disposal flow chart of high-level radwaste

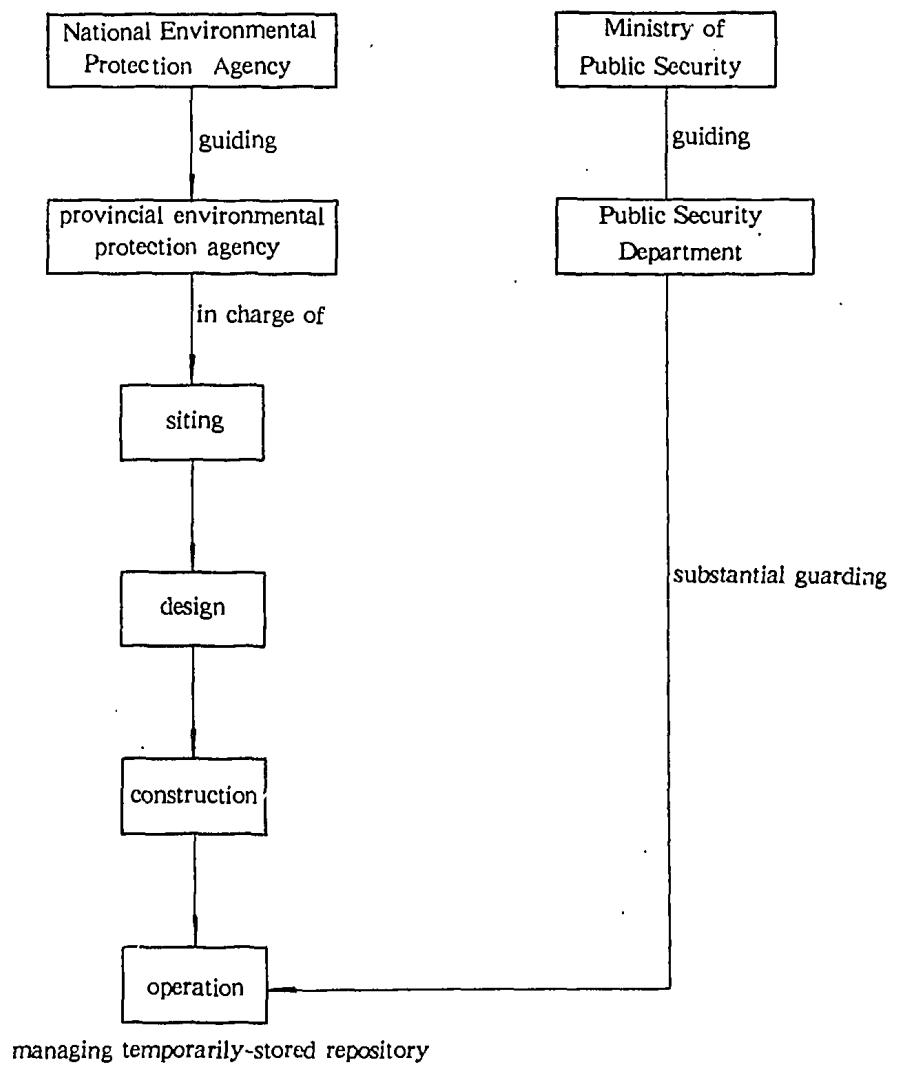


Fig. 6 Management flow chart of radwaste from nuclear technology application

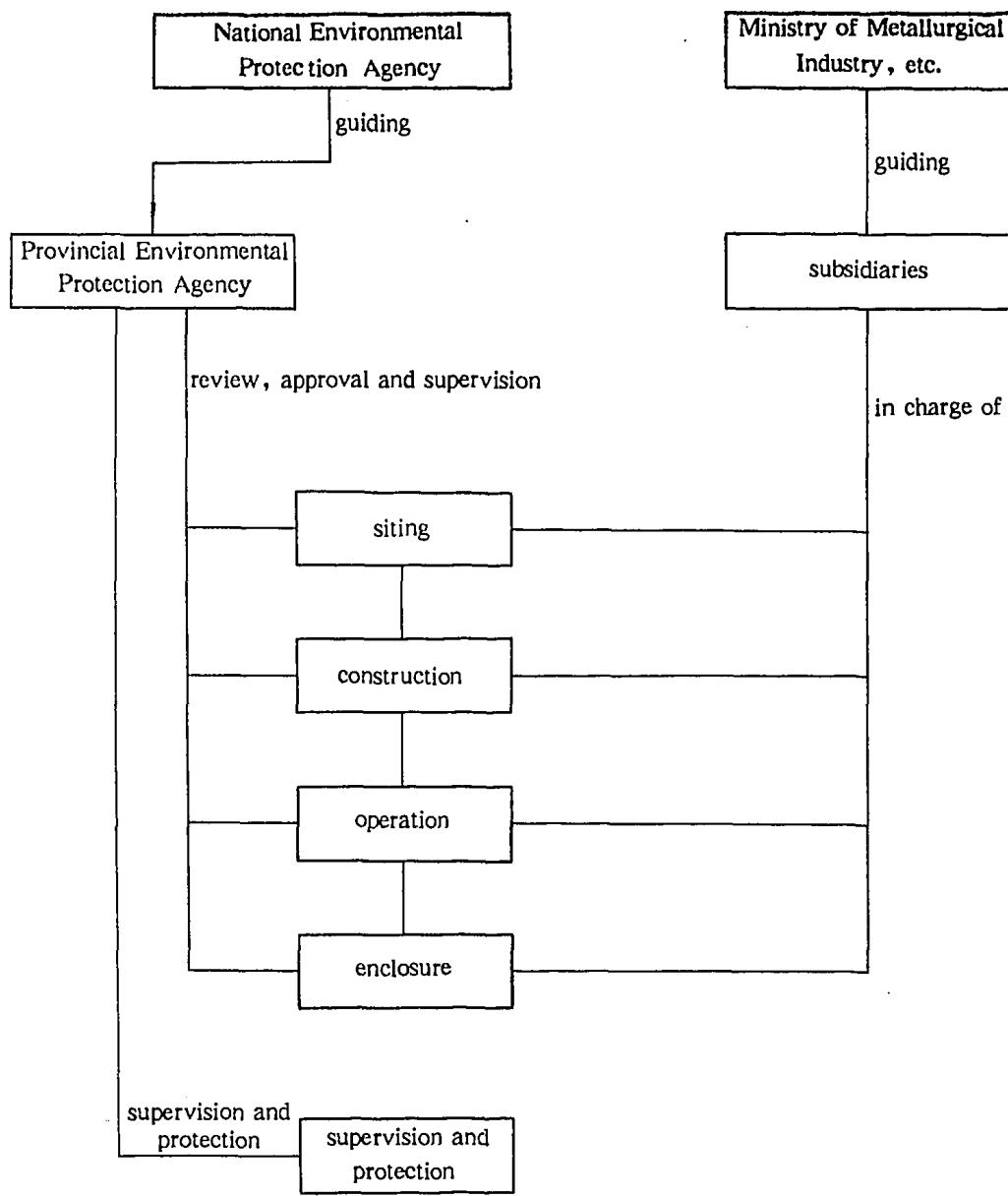


Fig. 7 Management flow chart of tailings dam for non-uranium deposit

## References

- [1] United Nations Conference On Environment and Development Rio de Janeiro, 3—14 June 1992, Agenda 21.
- [2] NEPA, China National Environmental Protection Agency, Environmental Policy of China on Intermediate-and Low-Radwaste Disposal, June 24, 1992.
- [3] Zhao Yamin, Requirements of Environmental Protection for High-level Radwaste deep geological disposal (to be passed)
- [4] NEPA, China National Environmental protection Agency, Management Criteria for Radwastes from Nuclear Technology Application, July 16, 1987.

# NON-DESTRUCTIVE MEASUREMENTS OF NUCLEAR WASTES

## VALIDATION AND INDUSTRIAL OPERATING EXPERIENCE

A. SAAS  
CEA, FRANCE  
Eric TCHEMITCIIEFF  
SGN, FRANCE

### ABSTRACT

After a short survey of the means employed for the non-destructive measurement of specific activities ( $\gamma$  and X-ray) in waste packages and raw waste, the performances of the device and the ANDRA requirements are presented.

The validation of the  $\gamma$  and X-ray measurements on packages is obtained through determining, by destructive means, the same activity on coring samples. The same procedure is used for validating the homogeneity measurements on packages (either homogeneous or heterogeneous).

Different operating experiences are then exposed for several kinds of packages and waste.

Up to now, about twenty different types of packages have been examined and more than 200 packages have allowed the calibration, validation, and control.

### 1. Introduction

Characterization and control assays performed on request of the waste producer, waste manager or safety authorities, include the determination of mass activity either in raw waste or in conditioned or immobilized waste packages.

For the purpose, the Service de Caractérisation, d'Evaluation des Confinements et d'Analyses (SCECA), through the Laboratoire d'Expertise et de Caractérisation des Confinements (LECC) and the Section d'Analyses des Effluents et Déchets (SAED), has developed, for its own use and for producers, some mobile devices suitable for all waste production, regardless of the type of waste, matrix, and package.

Operating experience is available on several dozen packages:

- CEN—Cadarache packages (homogeneous and heterogeneous cemented waste),
- STE<sub>3</sub> bitumen packages,
- STEL bitumen packages, and
- SSM packages (ANDRA).

This paper briefly discusses the means employed, the French requirements for radioactive homogeneity and mass activity measurement; then describes the validation tests required for all types of waste and finally presents some examples of industrial measurements.

### 2. Examination Means

The means used comprise mainly:

- a "high purity Germanium" gamma detector (range: 10 keV to 10 MeV; resolution: 1 keV at

- 122 keV and 2 keV at 1.33 MeV),
- a "low-energy germanium" gamma detector (range: 3 keV to 1 MeV; resolution: 230 eV at 5.9 keV and 540 eV at 122 keV),
  - a 4096-channel analyzer,
  - a microcomputer,
  - specialized spectrum analysis software,
  - an optical detector alignment system,
  - accessories for liquid nitrogen,
  - lead shielding for collimation,
  - a radioactive sealed source for calibration, and
  - a turnable for waste packages (permissible load = 10 tons; rotation speed = 1 revolution per minute).

*All these examination means are mobile* and can thus be used to perform measurements directly in the producer's facilities, reducing measurement costs by eliminating the need to ship radwaste packages to the laboratory.

The turnable is specifically used for heterogeneous packages and to test the containment properties of overpacks and concrete shells.

Table 1 shows the application scope and the main device performance specifications.

### 3. French Homogeneity and Mass Activity Determination Requirements for Radwaste to be Delivered to ANDRA

Current ANDRA regulations define homogeneity requirements, embedding thresholds, and the maximum permissible activities for the main isotopes.

#### Waste package homogeneity

The homogeneity of a homogeneous radwaste package is defined by a maximum mass concentration difference of  $\pm 25\%$  from the mean activity.

To be acceptable, the mass activity in any 100-litre volume inside a heterogeneous waste package must not exceed one-fifth of the total activity.

#### Embedding threshold

This is the mass activity above which waste must be conditioned; two types of thresholds are specified:

- a total emission threshold:  
e. g.  $\alpha = 5 \times 10^{-3}$  Ci/t  
 $\beta\gamma = 1$  Ci/t;
- individual thresholds for each isotope with a half-life exceeding 180 days.

#### Maximum permissible activity

This activity is defined for each radionuclide;

- e. g. 1300 Ci/t of  $^{60}\text{Co}$   
130 Ci/t of  $^{137}\text{Cs}$   
total  $\alpha = 0.1$  Ci/t.

The current requirements on measurements levels for raw waste, packages, and leachates are shown in Table 2 for homogeneous waste.

#### 4. Validation Tests on Measurements

These validation tests were carried out concurrently on raw waste and conditioned waste; the validation concerns both  $\beta\gamma$  and  $\alpha$ ( $^{241}\text{Am}$ ) emitters and emission ratios for pure  $\beta$  and  $\alpha$  emitters, e. g.  $^{60}\text{Co}/^{63}\text{Ni}$ ;  $^{137}\text{Cs}/^{90}\text{Sr}$ ;  $^{241}\text{Am}/\text{total } \alpha$ .

##### 4.1 Validation on raw waste

Two objectives were assigned:

- validation of sampling and of the destructive techniques necessarily used for pure  $\beta$  emitters in order to define the ratios; and
- sample optimization for measurements (mass, measurement representativity, uncertainties).

These aims are illustrated by the results shown in Tables 3 and 4. In Table 3, graphite is used as an example; non-destructive measurements on about 30 samples were used to validate:

- the minimum number of samples to examine (3),
- the dissolution technique, and
- the definition of the main ratios.

In Table 4, the example chosen illustrates the optimization of sample mass for ion exchange resins and the processing of primary samples.

The following remarks are applicable to these examples:

- for each type of waste, the preparation and dissolution techniques of raw samples must be validated; and
- the optimum mass for raw solid waste is about 10 grams.

##### 4.2 Validation of conditioned waste packages

Several steps were necessary to validate the non-destructive measurements on packages:

- comparison of measurements on the package and its core samples using the same technique;
- comparison of measurements on the package and the samples (taken from the coring samples) by non-destructive means;
- comparison of non-destructive and destructive results; and
- comparison of different measurement procedures (immobilized package, rotating package, different detectors).

##### Comparison between the package and its core sample

By coring without water, the laboratory can take several samples from the package, to control the activity level and distribution, and to perform different tests (compression, leaching, etc.).

An example of activity comparison on a cemented concentrates package is given in Table 5. These measurements were carried out by non-destructive means. The good correlation of measurements can be seen.

##### Comparison between the package, the core samples and their slices

The purpose of these non-destructive validation tests is to confirm that the self-shielding coefficients, the geometry and the scale effect allow the technique to be used for any test of

characterization, control, and inspection.

An example of this type of validation is given in Table 6. These results come from control measurements on cemented concentrates from a nuclear power plant. The deviations between the different measurements do not exceed 10 to 15%.

#### Comparison between destructive and non-destructive measurements

The aim here is to allow a complete cross-checking of measurements on mass activities of all the isotopes concerned by the requirements on conditioned waste:  $\beta\gamma$ , pure  $\beta$ , and  $\alpha$  emitters.

The example in Table 7 shows the results of control measurements on an old 700-litre package (cemented concentrates, weighing 1.4 ton). The measurements allow a good evaluation of the mass activity of this package.

#### Comparison between the different types and means of non-destructive measurements

Although the calibrations and validations above allow a good cross-checking of the measurements, the influence of the measurement mode on heterogeneous packages must be assessed.

Table 8 shows the results given by measurements on steady and rotating packages, both homogeneous and heterogeneous. It is clear that heterogeneous packages must be rotated to measure them. With overcoated packages, the containment must also be tested by measuring possible diffusion of activity in the overcoating.

The use of several types of detectors and the need for evaluating  $\alpha$  activity through measuring  $^{241}\text{Am}$  with low-energy detector also call for validation of this technique.

An example is given in Table 9; the cross-checking of  $^{241}\text{Am}$  measurements by a  $\gamma$ -emitter such as  $^{137}\text{Cs}$  validates this type of measurement.

These tests, calibrations, destructive and non-destructive measurements allowed validation of the device employed for determining the mass activity of packages and raw waste in compliance with ANDRA requirements.

The second step involves industrial operating experience.

## 5. Industrial Measurement Operating Experience

The measurements and operating experience presented below concern:

- homogeneity measurements on homogeneous and heterogeneous packages to be shipped to the surface storage site,
- determination of conversion tables for decommissioning waste,
- water filter measurements in routine operation before immobilization in concrete shells,
- special waste measurements: after incident, or waste waiting for transportation, old waste removed from storage, and
- measurement for experimental operation when starting new facilities.

### 5.1 Homogeneity measurements on packages before shipping to surface storage sites

These measurements are mainly carried out at the Cadarache Nuclear Research Center but also for control. Two examples are given: the first one concerns a package from Cadarache (homogeneous waste); Table 10 illustrates the technique used and the results obtained for a  $\beta\gamma$

emitter ( $^{137}\text{Cs}$ ) and an  $\alpha$  emitter ( $^{241}\text{Am}$ ), for the latter radionuclide, Fig. 1 shows the  $^{241}\text{Am}$  spectrum obtained by low-energy detector for one measurement point.

The second example concerns a heterogeneous package; Table 11 shows an application of the regulation that limits the activity inside any 100-litre volume to one-fifth of total activity. The percentage of relative activity (100 %) corresponds to 5 % of total mass activity.

## 5. 2 Conversion tables for decommissioning waste

In addition to actual dismantled waste, decommissioning generates technological waste (gloves, cotton, etc.). To ensure rational operation, routine controls are performed and it is very useful to define conversion tables relating routine dose rate measurements to total activity and typical spectrum.

An industrial example is given below, with all its steps:

- the first step consists in non-destructive measurements on several series of packages issued from the different zones of the facility being decommissioned, (Table 12);
- the second step is to determine the typical spectra and the different waste characteristics (density, dose rate), (Table 13); and
- the last step consists in computing the conversion coefficients applicable to each type of waste (Table 14).

These steps are carried out as the decommissioning work progresses.

## 5. 3 Routine measurements on water filters before immobilization in concrete shells

Water filters from nuclear power plants are waste with activities varying over a wide range, from a few mCi to several tenths of Curies per filter.

Although this kind of filter has a typical spectrum in normal operation, variations are observed during plant starting or in case of serious cladding failure; it is then necessary to measure filter activity before immobilizing them in concrete shells.

An example of routine measurement is given in Table 15. This example shows the activity dispersion according to filter location, as well as the fluctuations of isotopic ratios:

- e. g.  $^{134}\text{Cs}/^{137}\text{Cs}$ :  $1.46 \times 10^{-2}$   
 $^{60}\text{Co}/^{137}\text{Cs}$ :  $1.74 \times 10^{-2}$  to  $1.7 \times 10^{-3}$   
 $^{60}\text{Co}/^{125}\text{Sb}$ : 0.195 to 1.567.

## 5. 4 Measurements on special waste

The number of non-destructive measurement needed for this type of waste increases with the need for storage and the requests to ANDRA for special packages.

Three examples illustrate this operating experience:

- measurements on a settling tank from a nuclear power plant: this vessel was placed in a container for failed assemblies, surrounded by concrete shielding. Table 16 shows the results of  $\gamma$  and low-energy  $\gamma$  measurements;
- measurements on a control rod after replacement: Figure 2 shows the contents of this package. Table 17 shows both the measured spectrum including some minor radionuclides such as  $^{121}\text{Sb}$ , and the activity of the two major isotopes:  $^{58}\text{Co}$  and  $^{60}\text{Co}$  (activity between 3,800 and 4,000 Ci); and
- measurements on packages to be removed from storage because of radium  $\alpha$ -activity. In

addition to the overall activity per package, it was necessary to measure the distribution in the drum and in the absorbents placed at the top and bottom of the drum. Table 18 shows an example of measurement for this type of package.

### 5.5 Industrial measurements performed when starting new facilities

Non-destructive measurements are performed on bitumized packages during the experimental operation periods.

The industrial targets are:

- validation of the activity evaluations made by the operator from measurements of raw waste;
- validation of the industrial measurement system based on dose rate measurement; and
- verification that the results comply with the specifications and guaranteed process parameters.

Table 19 shows an example of the measurements performed and of the evaluations given by the operator. The deviations between the evaluations, the full-scale non-destructive measurements and the dosimetries ranged from 15 to 20 % for packages containing 60 to 400 Ci of  $\beta\gamma$  emitters.

## Conclusions

These validation results and industrial measurements show that the measurements performed by the Laboratoire d'Expertise et de Caractérisation des Confinements comply with current ANDRA specifications for homogeneity and for mass activity of packages. Nevertheless, the only  $\alpha$ -emitter that can be routinely measured at the present time is  $^{241}\text{Am}$ . This determination will be used for relating with neutron measurements (active or passive) and for non-destructive validation of all the devices needed to assess mass activity for  $\beta$ ,  $\gamma$  and  $\alpha$  emitters.

Non-destructive  $\beta\gamma$  measurements will remain necessary to determine pure  $\beta$  emitters and validate the destructive techniques employed for this type of isotope.

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Table 1 Application fields and performance

### 1. Application fields

#### Complete packages:

- Drums
- Shells
- Homogeneous or heterogeneous wastes in different matrices

#### Samples:

- Core-samples, filters, control rods, unconditioned dismantling wastes, graphite, raw wastes, wastes with radium, etc.

### 2. $\beta\gamma$ - X-ray activity · performance

$\beta\gamma$ : up to 1000 Ci/package or raw waste

X-ray: up to 50 Ci/package or raw waste

### 3. Detection limits

$\beta\gamma$  activity:  $10^{-3}$  to  $10^{-4}$  Ci/t per emitter

X-ray: ( $^{241}\text{Am}$ ) :  $10^{-2}$  to  $10^{-3}$  Ci/t

Table 2 Sensitivity and detection limit required for activity measurements for raw and conditioned wastes

Specification	Activity level	Emitter	Detection limit	Uncertainty of results
Embedding limit	$10^{-3}$ Ci/t	$\alpha$	$5 \times 10^{-6}$ Ci/m <sup>3</sup>	50%
Acceptability limit	0.1 Ci/t	$\alpha$	$5 \times 10^{-7}$ Ci/m <sup>3</sup>	10~20%
Maximum limit	1 Ci/t	$\alpha$	$5 \times 10^{-6}$ Ci/m <sup>3</sup>	5~10%
Embedding limit	0.1 Ci/t	$\beta\gamma$	$5 \times 10^{-7}$ Ci/m <sup>3</sup>	50%
Embedding limit	1 Ci/t	$\beta\gamma$	$5 \times 10^{-6}$ Ci/m <sup>3</sup>	10~20%
Acceptability limit (medium)	10 to 20 Ci/t	$\beta\gamma$	$10^{-4}$ Ci/m <sup>3</sup>	5~10%
Maximum limit	1000 Ci/t	$\beta\gamma$	$10^{-2}$ Ci/m <sup>3</sup>	5%
Acceptability limit	2 Ci/t	H <sub>3</sub>	$5 \times 10^{-7}$ Ci/m <sup>3</sup>	20%
Embedding limit	$10^{-4}$ Ci/t	$\gamma$ long half-life	( $10^{-10}$ - $10^{-11}$ ) Ci/m <sup>3</sup>	50%
Acceptability limit	$10^{-1}$ Ci/t	$\gamma$ long half-life	( $10^{-7}$ - $10^{-8}$ ) Ci/m <sup>3</sup>	10~20%
Embedding limit	$10^{-3}$ Ci/t	$\beta$ long half-life	$10^{-9}$ Ci/m <sup>3</sup>	50%
Acceptability limit	$10^{-1}$ Ci/t	$\beta$ long half-life	( $10^{-7}$ - $10^{-8}$ ) Ci/m <sup>3</sup>	10~20%
Maximum limit	10 Ci/t	$\beta$ long half-life	( $10^{-5}$ - $10^{-6}$ ) Ci/m <sup>3</sup>	5~10%

Table 3 Graphite sample measurements

Non-destructive  $\beta\gamma$  activity measurements

<sup>60</sup> Co: 1 to 12	kBq/g ( $0.3 - 3.2 \times 10^{-1}$ Ci/t)
<sup>133</sup> Ba: 0.05 to 0.1	kBq/g ( $0.15 - 0.3 \times 10^{-2}$ Ci/t)
<sup>134</sup> Cs: 0.01	kBq/g ( $0.3 \times 10^{-3}$ Ci/t)
<sup>137</sup> Cs: 0.01 to 0.15	kBq/g (( $0.03 - 4.5$ ) $\times 10^{-3}$ Ci/t)
<sup>154</sup> Eu: 0.4 to 0.8	kBq/g (( $1.2 - 2.4$ ) $\times 10^{-2}$ Ci/t)

Destructive measurements

<sup>3</sup> H: 340 - 400	kBq/g (9.2 - 10.8 Ci/t)
<sup>14</sup> C: 6 - 25	kBq/g (0.2 - 0.7 Ci/t)
<sup>63</sup> Ni: 1 - 7.5	kBq/g (0.3 - 2.0 $\times 10^{-1}$ Ci/t)
<sup>93m</sup> Nb: < 0.1	kBq/g (< $0.3 \times 10^{-2}$ Ci/t)
<sup>36</sup> Cl: 0.4 - 1.5	kBq/g (( $1.2 - 4.5$ ) $\times 10^{-2}$ Ci/t)
<sup>60</sup> Co: 2 - 15	kBq/g (( $0.6 - 4.0$ ) $\times 10^{-1}$ Ci/t)
<sup>133</sup> Ba: 0.05 - 0.2	kBq/g (( $0.15 - 0.6$ ) $\times 10^{-2}$ Ci/t)
<sup>134</sup> Cs: 0.01 - 0.1	kBq/g (( $0.3 - 3$ ) $\times 10^{-3}$ Ci/t)
<sup>137</sup> Cs: 0.01 - 0.1	kBq/g (( $0.3 - 3$ ) $\times 10^{-3}$ Ci/t)
<sup>154</sup> Eu: 0.4 - 0.8	kBq/g (( $1.2 - 2.4$ ) $\times 10^{-2}$ Ci/t)
<sup>155</sup> Eu: 0.15 - 0.4	kBq/g (( $0.4 - 1.2$ ) $\times 10^{-2}$ Ci/t)

Ratios for different isotopes

<sup>14</sup> C/total $\beta\gamma$ activity	: $1.7 \pm 0.7$
<sup>63</sup> Ni/total $\beta\gamma$ activity	: $0.5 \pm 0.1$
<sup>63</sup> Ni/ <sup>60</sup> Co	: $0.58 \pm 0.16$

Table 4 Validation and optimization for sampling

1. Destructive and Non-destructive measurements for ion exchange resins (IER)

Radionuclide	Total sample (non-destructive)	Sample fractions (destructive) 10 g	Sample fractions (destructive) 13 g
<sup>54</sup> Mn	25.5 ± 0.8	17.7 ± 1.3	37.6 ± 2.6
<sup>58</sup> Co	28.0 ± 0.7	19.0 ± 6.8	40.0 ± 8.2
<sup>60</sup> Co	54.0 ± 7.6	38.0 ± 2.3	79.3 ± 4.2
<sup>110m</sup> Ag	52 ± 1.5	62.3 ± 4.8	63.0 ± 4.8

2. Validation of sampling to optimize the measurements (destructive analysis)

Dry weight	<sup>54</sup> Mn	<sup>60</sup> Co	<sup>110m</sup> Ag
6.4 g	36.6 ± 4.4	73.4 ± 6.2	71.0 ± 8.0
12.6 g	33.5 ± 3.3	76.4 ± 5.8	68.1 ± 6.8
14.6 g	34.5 ± 3.6	76.8 ± 5.5	64.8 ± 6.5
22.4 g	33.7 ± 3.0	72.2 ± 4.9	63.2 ± 4.9
31.9 g	32.2 ± 2.6	72.5 ± 4.7	64.2 ± 5.5
51.2 g	30.4 ± 2.6	66.2 ± 4.2	56.7 ± 4.6
Moyenne	33.5 ± 3.0	72.9 ± 5.0	64.7 ± 6.0
Deviation from Mean:			
Maximum	1.09	1.05	1.10
Minimum	0.91	0.91	0.88

Table 5 Comparison between full scale package and core sample  
(activity relative to full scale package in mCi)

Radionuclide	Core sample	Full scale package
<sup>60</sup> Co	0.34 ± 0.051	0.30 ± 0.046
<sup>134</sup> Cs	0.078 ± 0.012	0.080 ± 0.012
<sup>137</sup> Cs	71 ± 11	80 ± 12

Table 6 Non-destructive measurement validation from different size of samples  
(concentrates in cement)

Radionuclide		Full scale package (220 l - 370 kg)	Core sample (800×60 mm - 15 kg)	Leaching sampling (80×80 mm - 0.6 kg)
<sup>54</sup> Mn	Bq mCi	(4.2 ± 0.6)×10 <sup>8</sup> 11.4 ± 1.6	(3.1 - 3.2)×10 <sup>8</sup> 8.4 - 8.8	(4.5 - 4.8)×10 <sup>8</sup> 12.3 - 12.9
<sup>60</sup> Co	Bq mCi	(1.6 ± 0.2)×10 <sup>9</sup> 43.2 ± 5.4	(1.35 - 1.39)×10 <sup>9</sup> 36.5 - 37.4	(1.51 - 1.53)×10 <sup>9</sup> 40.8 - 41.4
<sup>134</sup> Cs	Bq mCi	(1.1 ± 0.2)×10 <sup>8</sup> 3.0 ± 0.5	(9.25 - 9.62)×10 <sup>7</sup> 2.5 - 2.6	1.13×10 <sup>8</sup> 3.05
<sup>137</sup> Cs	Bq mCi	(9.3 ± 1.4)×10 <sup>8</sup> 25.1 ± 3.8	(9.10 - 9.17)×10 <sup>8</sup> 24.6 - 24.8	(1.03 - 1.05)×10 <sup>9</sup> 27.8 - 28.4
Total	Bq mCi	(3.1 ± 0.5)×10 <sup>9</sup> 84.0 ± 12	(2.70 - 2.75)×10 <sup>9</sup> 72.0 to 74 ± 7	(3.1 - 3.2)×10 <sup>9</sup> 84 ± 8 to 86 ± 8

Table 7 Comparison between destructive and non-destructive measurements  
(activity relative to full scale package in mCi)

Radionuclide	Full scale package	Core sample	Sample 5 - 15 g	Test samples ≈10 g
<sup>60</sup> Co	0.30 ± 0.046	0.34 ± 0.051	0.35 ± 0.05	0.30 ± 0.05
<sup>134</sup> Cs	0.080 ± 0.012	0.078 ± 0.012	0.081 ± 0.012	0.075 ± 0.015
<sup>137</sup> Cs	80 ± 12	71 ± 11	25 ± 12	78 ± 10
<sup>241</sup> Am	1.4 ± 0.2	—	1.3 ± 0.1	1.6 ± 0.2
<sup>90</sup> Sr	—	—	—	0.19 ± 0.03

Table 8 Type of measurements for heterogeneous and homogeneous packages

Homogeneous package measurement

Radionuclide	Static measurement	Measurement with turntable
<sup>60</sup> Co	0.30 ± 0.046	0.32 ± 0.048
<sup>134</sup> Cs	0.08 ± 0.012	0.09 ± 0.016
<sup>137</sup> Cs	80 ± 12	79.7 ± 12

Heterogeneous package measurement

Radionuclide	Static measurement	Measurement with turntable
<sup>60</sup> Co	6.70 ± 0.6	7.20 ± 0.6
<sup>134</sup> Cs	1.5 ± 0.1	1.9 ± 0.2
<sup>137</sup> Cs	15.3 ± 1.5	37.9 ± 4

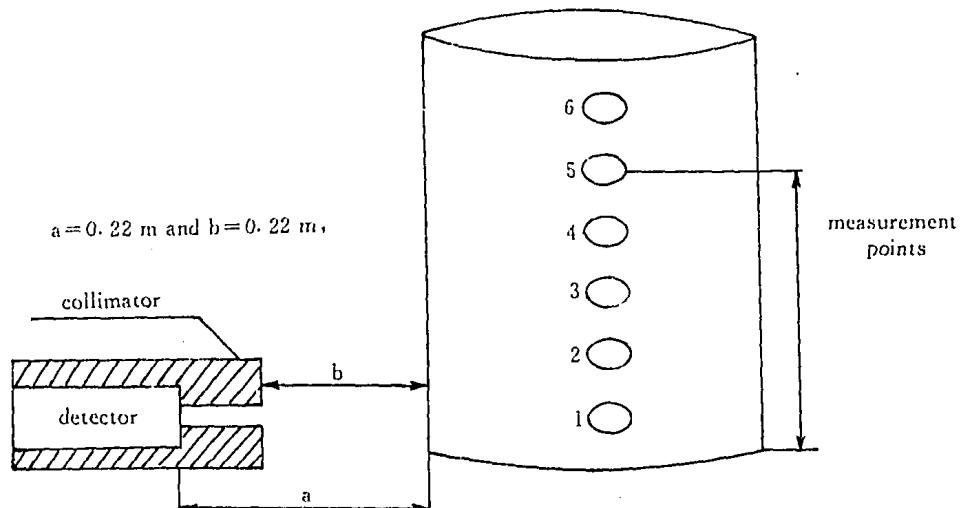
Table 9 Equipment and sample comparison  
 Comparison of measurements by different devices; ( $^{241}\text{Am}$ ) homogeneous  
Package measurement

Samples (packages)	$\gamma$ probe	X-ray probe
1	$1.4 \times 10^3 \pm 300$	$1.6 \times 10^3 \pm 350$
2	$2.7 \times 10^4 \pm 6000$	$2.5 \times 10^4 \pm 5500$

Comparison of measurements from different types of samples (MBq/drum)

Type of samples	$^{241}\text{Am}$	$^{137}\text{Cs}$
Full scale package (1.4 t)	$1.4 \times 10^3$	$2.9 \times 10^3$
Core sample (16 kg)	Non measured	$2.66 \times 10^3$
Section of core sample (1 kg)	$0.9 \times 10^3$	$2.84 \times 10^3$
Destructive measurement of sample (10 g)	$1.6 \times 10^3$	$2.80 \times 10^3$

Table 10 Results for homogeneity control (7001 PACKAGE No. 1901)



Measurement points	Height (m)	Relative activity (%)	
		$^{137}\text{Cs}$	$^{241}\text{Am}$
1	0.15	$82 \pm 8$	$92 \pm 9$
2	0.30	$76 \pm 8$	$78 \pm 8$
3	0.45	$87 \pm 9$	$84 \pm 8$
4	0.60	$100 \pm 10$	$100 \pm 10$
5	0.75	$91 \pm 9$	$79 \pm 8$
6	0.90	$97 \pm 10$	$79 \pm 8$

Table 11  $\gamma$  scanning of heterogeneous package  
(% of relative activity)

Measurement points	$^{60}\text{Co}$	$^{137}\text{Cs}$
Surface	100	100
Medium	250	510
Bottom	30	15

Table 12 Measurements of dismantling waste:  
raw waste measurements

Reference of drums	Routine control measurements	$^{22}\text{Na}$ kBq	$^{54}\text{Mn}$ kBq	$^{60}\text{Co}$ kBq	$^{134}\text{Cs}$ kBq	$^{137}\text{Cs}$ kBq
4751	Undetectable	—	—	< 37	—	< 37
4752	1700	—	—	$20190 \pm 1700$	—	$2375 \pm 236$
4753	700	$7.0 \pm 1.4$	$337 \pm 31$	$365 \pm 31$	$98 \pm 10$	$14393 \pm 1400$
4754	700	—	—	$6016 \pm 52$	—	$2915 \pm 280$
4756	3300	—	$2527 \pm 235$	$3197 \pm 277$	$673 \pm 68$	$101800 \pm 9500$

Table 13 Dismantling waste  
density — dose rate — % of activity for different isotopes

Reference of drums Checking cart	4751	4752	4754	4753	4756
Density	Not measured	$0.22 \text{ g/cm}^3$	$0.28 \text{ g/cm}^3$	$0.24 \text{ g/cm}^3$	$0.24 \text{ g/cm}^3$
Origin	Hut changing room	Decontamination	Decontamination	Reactor containment	Reactor containment
Maximum contact dose rate	< 0.1 mrad/h	40 mrad/h at mid-height	4 mrad/h at mid-height 10 mrad/h at bottom	4 mrad/h at mid-height 10 mrad/h at bottom	4 mrad/h at mid-height 10 mrad/h at bottom
Relative activity					
$^{22}\text{Na}$	—	—	—	0.05%	
$^{54}\text{Mn}$	—	—	—	2.20%	2.3%
$^{60}\text{Co}$	50%	80%	68%	2.50%	3.1%
$^{134}\text{Cs}$	—	—	—	0.60%	0.6%
$^{137}\text{Cs}$	50%	10%	32%	94.65%	94.0%

Table 14 Dismantling waste  
(conversion table; count to activity in kBq)

Type of wastes	Conversion factors
Decontamination	13.3 kg $\pm$ 20% FOR 1 count per second
Reactor barrier	27.2 kBq $\pm$ 20% FOR 1 count per second

Table 15 Process wastes (filters)  
(activity measurements: MBq/filter)

Sample number	$^{60}\text{Co}$	$^{125}\text{Sb}$	$^{154}\text{Eu}$	$^{134}\text{Cs}$	$^{137}\text{Cs}$
1	$42.2 \pm 7.0$	$106 \pm 25$	$89 \pm 21$	$99 \pm 14$	$6150 \pm 750$
2	$129 \pm 17$	$82.3 \pm 14.0$	$71 \pm 10$	$113 \pm 17$	$7400 \pm 900$
3	$190 \pm 32$	$950 \pm 210$	$1260 \pm 320$	$1273 \pm 177$	$84700 \pm 10700$
4	$218 \pm 35$	$1117 \pm 159$	$1140 \pm 285$	$1560 \pm 288$	$106300 \pm 10700$
5	$130 \pm 20$	$635 \pm 77$	$571 \pm 70$	$1143 \pm 158$	$76700 \pm 9700$

Table 16

Activity in a settling tank

Radionuclide	Activity in settling tank
$^{241}\text{Am}$	$30.0 \pm 7.5$
$^{125}\text{Sb}$	$20.6 \pm 4.0$
$^{154}\text{Eu}$	$40.0 \pm 6.0$
$^{134}\text{Cs}$	$42.0 \pm 6.0$
$^{137}\text{Cs}$	$4220 \pm 630$
$^{60}\text{Co}$	$13.6 \pm 2.0$

Activity calculated at measuring time (05/09/89)

Radionuclide	Activity (TBq)
$^{58}\text{Co}$	$69 \pm 12$
$^{60}\text{Co}$	$73 \pm 12$

Table 17 Control rod spectrum

Radionuclide	Activity (TBq)
$^{58}\text{Co}$	$69 \pm 12$
$^{60}\text{Co}$	$73 \pm 12$

Table 18

## Gamma-scanning of 200-liter drum for wastes containing radium

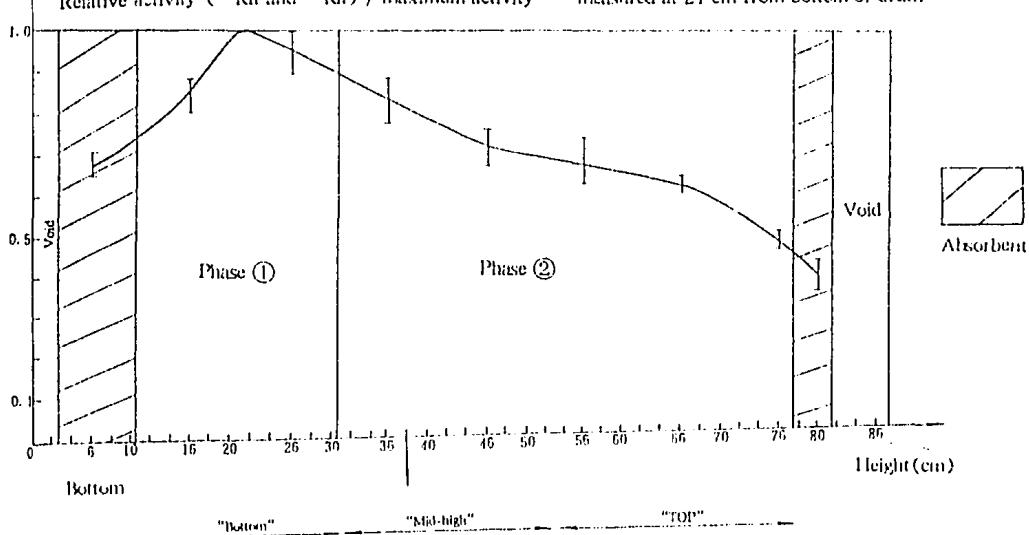
Relative activity ( $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ) / maximum activity — measured at 21 cm from bottom of drum

Table 19 Non-destructive measurements for startup of the STE3 bituminization facility at La Hague

Package number	$^{106}\text{Ru} + ^{106}\text{Rh}$ (Ci)		Total $\beta\gamma$ (Ci)		Dosimetry (rad/h)	
	LH	CAD	LH	CAD	LH	CAD
1099	26.01	23.0	64.1	58.5	45.0	$38.3 \pm 1.8$
1159	24.5	21.0	59.0	53.2	42	ND
992	147.1	122.1	340	314	102	$101.3 \pm 1.2$
993	141.0	118.0	333	292	ND	$96.7 \pm 2.9$

Non-destructive measurements (CAD)

Evaluation according to the operator's specifications (LH)

Validation of acceptance requirements (activity content in packages)

Fig. 1 Americium-241 Peak for the First Measurement Point for Package No. 1901

scale-2 K



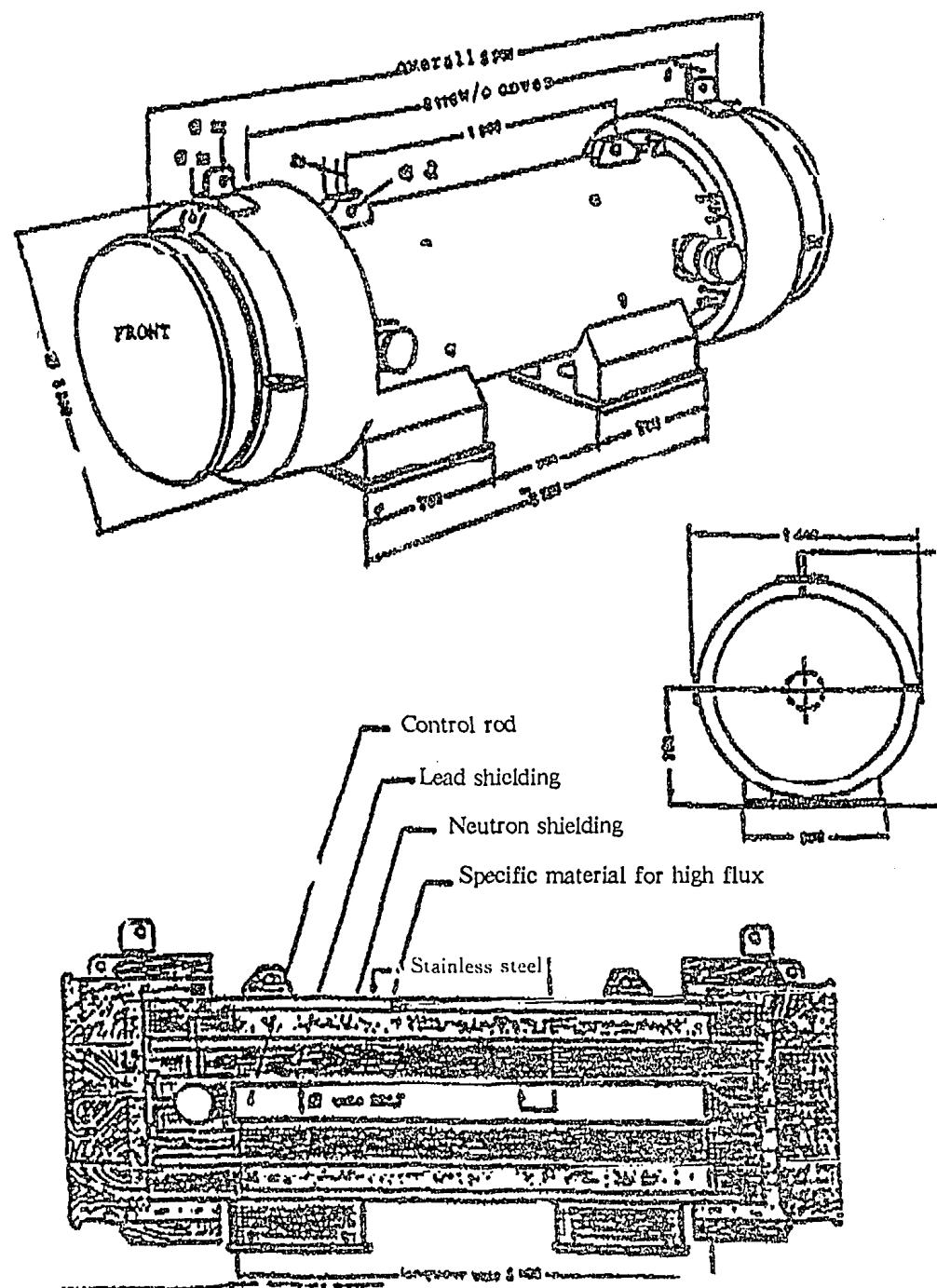


Fig. 2 Package Composition

# ISOTOPE HYDROLOGICAL CHARACTERISTICS OF GROUNDWATER OF PRESELECTED L/ILW DISPOSAL SITES

WANG Zhiming XIAO Feng LI Sen

Beijing Research Institute of Uranium Geology, China

## ABSTRACT

By studying the chemical compositions of the groundwater (deuterium, tritium, and stable isotope of oxygen-18, and the radioactive isotope of uranium) in the test area, the shallow groundwater (pore water) and the crevice-water in weathered bedrocks were successfully distinguished; the characteristics of groundwater outcrops in Ling'ao-Changwan area and the relationship between groundwater recharge, runoff, and discharge were discussed; and the relative age of different groundwater in the area was determined. The comprehensive analysis fairly conform to the conclusions obtained from the exploration activities. Therefore, a reliable scientific basis is provided to assess the preselected sites for disposal of low- and intermediate-level wastes(L/ILW).

### 1. Introduction

Ling'ao-Changwan area is located in the southeast coast of Dapeng Peninsula, Guangdong Province, with mountains behind and the sea in front, belonging to low mountain and hilly topography (an elevation of 200~400 m above sea level as a whole and the highest, 707.6 m). Most of mountain area and low-lying land is covered by thin eluvium, cliff debris, and diluvium, and few bedrock outcrops found in the area.

Regionally, complete strata from Cambrian to Quaternary are exposed, and intrusions are extensively distributed. The preselected site is located in the southeast wing of the anticline of Paiya Mountain, where only fine sandstone-siltstone intercalated with medium-sized sandstone and siliceous rocks of Guitou Member of Middle Devonian is found.

### 2. Hydrogeology

According to the topography, landform, and the conditions of surface water and groundwater development in the area, Ling'ao-Changwan area belongs to a similar hydrogeological unit (Fig. 1). Paiya Mountain-Hutou Mountain serves as the regional dividing range, and Beilong ridge, the local dividing range in the area. Thanks to the deep valley, close hydrographic network, and good discharge conditions of near-surface groundwater in the area, there is comparatively small amount of groundwater runoff. Pursuant to the difference in the aquiferous structure, the runoff distance of groundwater, pH value, TDS and the concentration of the main anion and cation in groundwater, the groundwater in the area can be recognized as three types: (1) Water in eluvium and cliff debris, which is the precipitation appearing in the surface after seeping into ground and migrating for a short distance; (2) Crevice-water in weathered rocks, which distributes around the local dividing range of Beilong ridge; and (3) Crevice-water in bedrocks in valleys and depression. The following hydrogeologic law is obeyed: much of the

precipitation flows along the slope surface and forms the surface water; some precipitation which filtrates into the ground is held up in eluvium and cliff debris for a period of time and then forms springs to compensate the surface water; some other precipitation goes into the weathered bedrock crevice, part of which flows southeastward to compensate laterally the bedrock crevice-water in valleys and depression, thus forming a mixture of two kinds of groundwater; finally, all kinds of groundwater discharge into the sea.

### 3. Composition of Hydrogen and Oxygen Isotopes in Groundwater

The composition of stable isotopes of hydrogen and oxygen in groundwater is an ideal tracer for studying the movement of natural water, which is useful in illustration of the mechanism of hydrogeological process. Analysis results of 12 samples collected from the area are shown in Table 1. By linear correlation analysis, the following linear relationship of local meteoric water is obtained:

$$\delta D (\text{\textperthousand}) = 6.68 \cdot \delta^{18}\text{O} + 7.5$$

The equation is roughly coincided with the linear equation of meteoric water in China. The decrease in slope and intercept of the line is mainly caused by certain degree of evaporation of surface water and groundwater.

It is showed in the figure of the relationship between  $\delta D$  and  $\delta^{18}\text{O}$  (Fig. 2) that (1) surface water and groundwater are recharged by precipitation; (2) near-surface groundwater is intensively exchanged; (3) crevice-water in weathered bedrocks has experienced certain water-rock interaction; and (4) there are certain mixing processes between near-surface groundwater and crevice-water in weathered bedrocks, i.e. the crevice-water in weathered bedrocks recharges the near surface groundwater.

### 4. Characteristics of Uranium Isotopes in Groundwater

Concentration of radioelements in groundwater, to some extent, indicates the hydraulic condition of groundwater in circulation, duration of interaction between groundwater and rock, and hydrogeochemical circulation. The analysis results of uranium isotopes collected from the groundwater of the area are shown in Table 2. Study on disequilibrium of uranium series in groundwater shows that there is a negative relationship between  $^{234}\text{U}/^{238}\text{U}$  ratio and uranium concentrations. Along the flowing direction, the farther the groundwater migrates from the recharge area, the more reductive the environment of groundwater is. Therefore, uranium in solution tends to move to the wall of aquifer. The main recharge area is the intermontane depression belt which is located at the upper part of the SE slope of Paiya Mountain, NW to the test area, and is formed by the anticline of Paiya Mountain. Groundwater filtrates through the paleo-weathered crust, and emerges at valleys encountered and forms descending spring to recharge surface water. The runoff direction of groundwater in the area is shown in Fig. 3.

### 5. Calculation for Relative Age of Groundwater

The test area is adjacent to Hong Kong.  $^3\text{H}$  data from Hong Kong observation stations published by the IAEA have been used for calculating the relative age of groundwater. Three groups of groundwater of different relative ages have been estimated. (1) Low  $^3\text{H}$  concentration water

formed before global nuclear test, the estimated age is older than 30 years, including bores ZK I -1, ZK II -1, and ZKH-1. This group of groundwater corresponds to bedrock crevice-water which has a low runoff rate; (2) crevice-water in weathered bedrocks with runoff duration of about 20 years; and (3) eluvium groundwater which is recharged by precipitation and river water in recent years. The above mentioned estimation can also be confirmed by the relationship between  $\text{HCO}_3^-$  and  $\delta^{18}\text{O}$  (Fig. 4).

## Conclusion

According to the comprehensive analysis based on the conditions of topography, landform and hydrogeology, and the characteristics of isotope hydrology, it is concluded that: Ling'ao-Changwan area lies in the same hydrogeological unit; Paiya Mountain-Hutou Mountain is the regional dividing range of the area; Beilong ridge is a local dividing range within the hydrogeological unit. The preselected site is located at the local dividing range and there is no risk of flood. The groundwater table is over 22 m and rocks of the preselected site are qualified in the property of engineering mechanics. All the above conditions coincide with the Regulation for Shallow Geological Disposal of Low- and Intermediate-Level Solid Radioactive Wastes (GB 9132-88). The preselected site is a suitable one for disposal of low- and intermediate-level solid radioactive wastes.

Table 1 Analytical Results of Isotope Composition in Groundwater in Ling'ao-Changwan Area

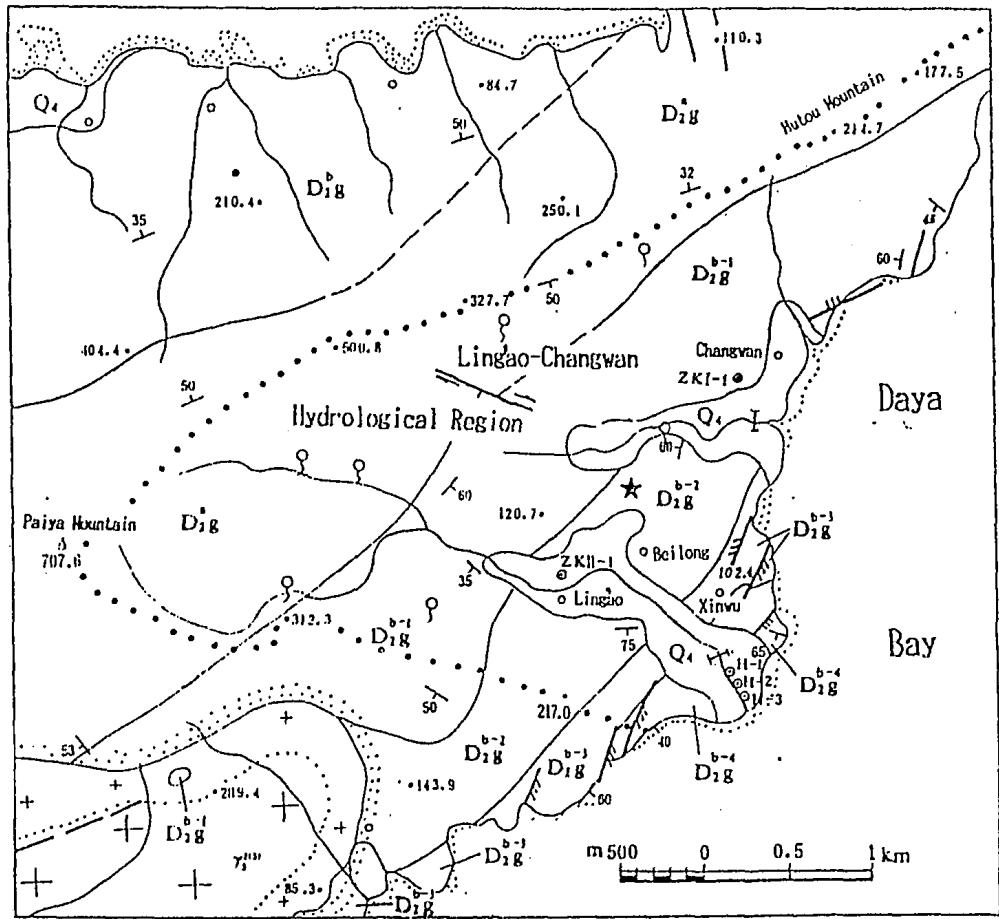
No.	Sampling Location & Water Table (m)	Sampling Depth(m)	Atmospheric Temp. (°C)	Water Temp. (°C)	$\delta D(\text{‰})$	$\delta^{18}\text{O}(\text{‰})$	$^3\text{H}(\text{TU})$	Time of Sampling
DY-1	ZK III -6 29.57	33.19	20.4	22.5	-50.5	-6.4	6.5	22/12/1991
DY-2	ZK III -5 27.96	29.20	23.0	22.5	-46.8	-6.2	7.8	22/12/1991
DY-3	ZK III -2 23.88	28.00	23.0	22.0	-45.2	-5.6	6.2	22/12/1991
DY-4	Civil well in Beilong 0.62		18.0	20.0	-48.8	-6.3	6.8	22/12/1991
DY-5	ZK I -1 8.24	12.24	19.0	22.9	-51.0	-6.2	2.2	23/12/1991
DY-6	ZKH-1 7.49	12.50	19.0	22.0	-48.0	-6.2	4.7	23/12/1991
DY-7	ZK II -1 7.79	11.79	23.0	22.0	-49.5	-6.3	5.1	23/12/1991
DY-8	Spring near the upper reach of Ling'ao stream		23.0	19.0	-47.4	-6.2	5.7	24/12/1991
DY-9	Spring near the north part of Beilong hill		20.5	18.5	-50.3	-6.4	7.5	24/12/1991
DY-10	Observation station of Changwan stream		19.0	18.0	-46.4	-6.0	5.3	24/12/1991
DY-11	Observation station of Ling'ao stream		21.0	19.5	-47.2	-6.1	6.0	25/12/1991
DY-12	Sea inlet of Ling'ao stream		27.0	19.0	-19.4	-0.8	5.6	25/12/1991

Note: 1. Tritium is analysed by Guangzhou Reserch Inst. of geology &amp; New Technology, Academia Sinica.

2. Stable isotope composition is analysed by Beijing Reserch Inst. of Uranium Geology.

Table 2 Analytical Results of Uranium Isotopes in Groundwater in Ling'ao-Changwan Area

No.	Sampling Location & Water Table (m)	Sampling Depth(m)	Atmospheric Temp. (°C)	Water Temp. (°C)	U (ppb)	$^{234}\text{U}/^{238}\text{U}$	Radioactive Activity		Time of Sampling
							$^{238}\text{U}(\text{mBq/l})$	$^{234}\text{U}(\text{mBq/l})$	
DY-1	ZK III -6 29.57	33.19	20.4	22.5	1.47	0.96	18.2	17.5	22/12/1991
DY-2	ZK III -5 27.96	29.20	23.0	22.5	1.15	0.96	14.3	13.7	22/12/1991
DY-3	ZK III -2 23.88	28.00	23.0	22.0	1.33	1.0	16.5	16.5	22/12/1991
DY-4	Civil well in Beilong 0.62		18.0	20.0	0.80	1.3	9.9	12.9	22/12/1991
DY-5	ZK I -1 8.24	12.24	19.0	22.9	0.91	1.1	11.3	12.4	23/12/1991
DY-6	ZKH-1 7.49	12.50	19.0	22.0	1.00	1.1	12.4	13.6	23/12/1991
DY-7	ZK II -1 7.79	11.79	23.0	22.0	1.45	0.91	18.1	16.5	23/12/1991
DY-8	Spring near the upper reach of Ling'ao stream		23.0	19.0	1.23	0.88	15.3	13.4	24/12/1991
DY-9	Spring near the north part of Beilong hill		20.5	18.5	1.00	1.2	12.4	14.9	24/12/1991
DY-10	Observation station of Changwan stream		19.0	18.0	0.87	0.94	10.8	10.1	24/12/1991
DY-11	Observation station of Ling'ao stream		21.0	19.5	0.88	1.0	10.9	10.9	25/12/1991
DY-12	Sea inlet of Ling'ao stream		27.0	19.0	2.97	1.2	35.8	44.2	25/12/1991



Legend

<b>Quaternary</b> <b>Guilou Formation of Devonian</b> <b>Yanshan cycle</b>	<table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="text-align: center; padding: 2px;"><b>Q<sub>t</sub></b></td><td style="text-align: left; padding: 2px;">Holocene Series</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>D<sub>ts</sub><sup>-4</sup></b></td><td style="text-align: left; padding: 2px;">Siltstone, muddy sandstone, and fine-grained sandstone intercalated with thin mud stone</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>D<sub>ts</sub><sup>-3</sup></b></td><td style="text-align: left; padding: 2px;">Medium-coarse grained sandstone intercalated with fine-grained sandstone and siltstone</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>D<sub>ts</sub><sup>-2</sup></b></td><td style="text-align: left; padding: 2px;">Fine-grained sandstone and siltstone intercalated with medium-grained sandstone and siliceous rock</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>D<sub>ts</sub><sup>-1</sup></b></td><td style="text-align: left; padding: 2px;">Coarse-medium grained sandstone intercalated with fine-grained sandstone and siltstone</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>D<sub>ts</sub><sup>0</sup></b></td><td style="text-align: left; padding: 2px;">Sandstone with gravel, sandstone, siltstone, and siliceous stone</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>D<sub>ts</sub><sup>-6</sup></b></td><td style="text-align: left; padding: 2px;">Conglomerate rock, sandstone, and siltstone</td></tr> </table> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="text-align: center; padding: 2px;"><b>y<sub>j</sub><sup>(1)</sup></b></td><td style="text-align: left; padding: 2px;">Granitite of Late Jurassic Period</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>/</b></td><td style="text-align: left; padding: 2px;">Actual and inferred geological boundary</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>\</b></td><td style="text-align: left; padding: 2px;">Compression fault</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>/\</b></td><td style="text-align: left; padding: 2px;">Compression-share fault</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>?</b></td><td style="text-align: left; padding: 2px;">Spring</td></tr> <tr> <td style="text-align: center; padding: 2px;">.....</td><td style="text-align: left; padding: 2px;">Boundary of dividing range</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>I—</b></td><td style="text-align: left; padding: 2px;">Observation station of surface water</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>ZK II-1</b></td><td style="text-align: left; padding: 2px;">Specified hydrological borehole and number</td></tr> <tr> <td style="text-align: center; padding: 2px;"><b>★</b></td><td style="text-align: left; padding: 2px;">Preselected site for disposal of low- and intermediate-level radwastes</td></tr> </table>	<b>Q<sub>t</sub></b>	Holocene Series	<b>D<sub>ts</sub><sup>-4</sup></b>	Siltstone, muddy sandstone, and fine-grained sandstone intercalated with thin mud stone	<b>D<sub>ts</sub><sup>-3</sup></b>	Medium-coarse grained sandstone intercalated with fine-grained sandstone and siltstone	<b>D<sub>ts</sub><sup>-2</sup></b>	Fine-grained sandstone and siltstone intercalated with medium-grained sandstone and siliceous rock	<b>D<sub>ts</sub><sup>-1</sup></b>	Coarse-medium grained sandstone intercalated with fine-grained sandstone and siltstone	<b>D<sub>ts</sub><sup>0</sup></b>	Sandstone with gravel, sandstone, siltstone, and siliceous stone	<b>D<sub>ts</sub><sup>-6</sup></b>	Conglomerate rock, sandstone, and siltstone	<b>y<sub>j</sub><sup>(1)</sup></b>	Granitite of Late Jurassic Period	<b>/</b>	Actual and inferred geological boundary	<b>\</b>	Compression fault	<b>/\</b>	Compression-share fault	<b>?</b>	Spring	.....	Boundary of dividing range	<b>I—</b>	Observation station of surface water	<b>ZK II-1</b>	Specified hydrological borehole and number	<b>★</b>	Preselected site for disposal of low- and intermediate-level radwastes
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Fig. 1 Schematic Hydrogeological Map of Ling'ao-Changwan Area

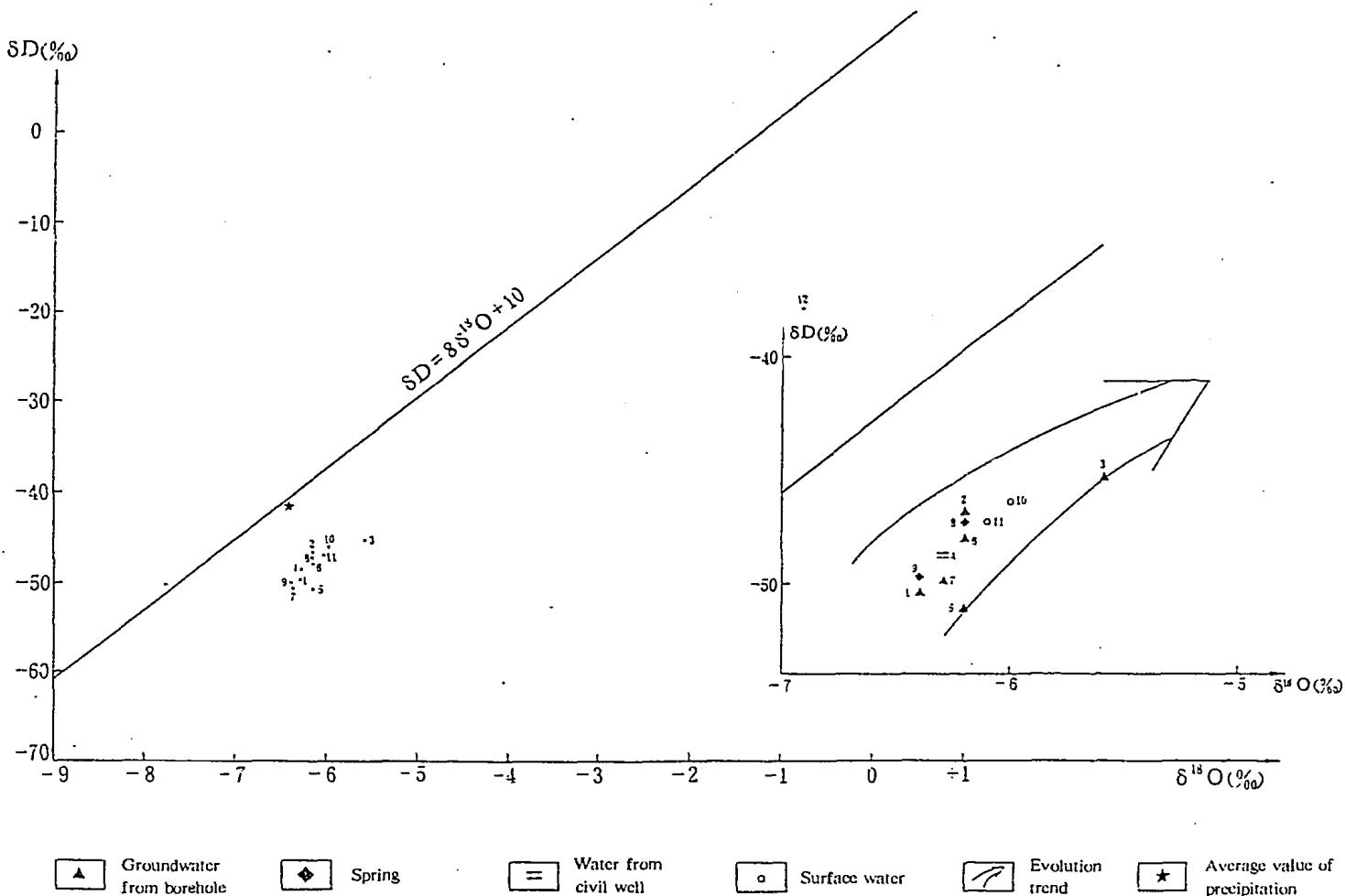


Fig. 2  $\delta\text{D}$ - $\delta^{18}\text{O}$  Relationship in Groundwater and Surface Water in Preselected Site  
for Disposal of Nuclear Wastes, Daya Bay Nuclear Power Plant

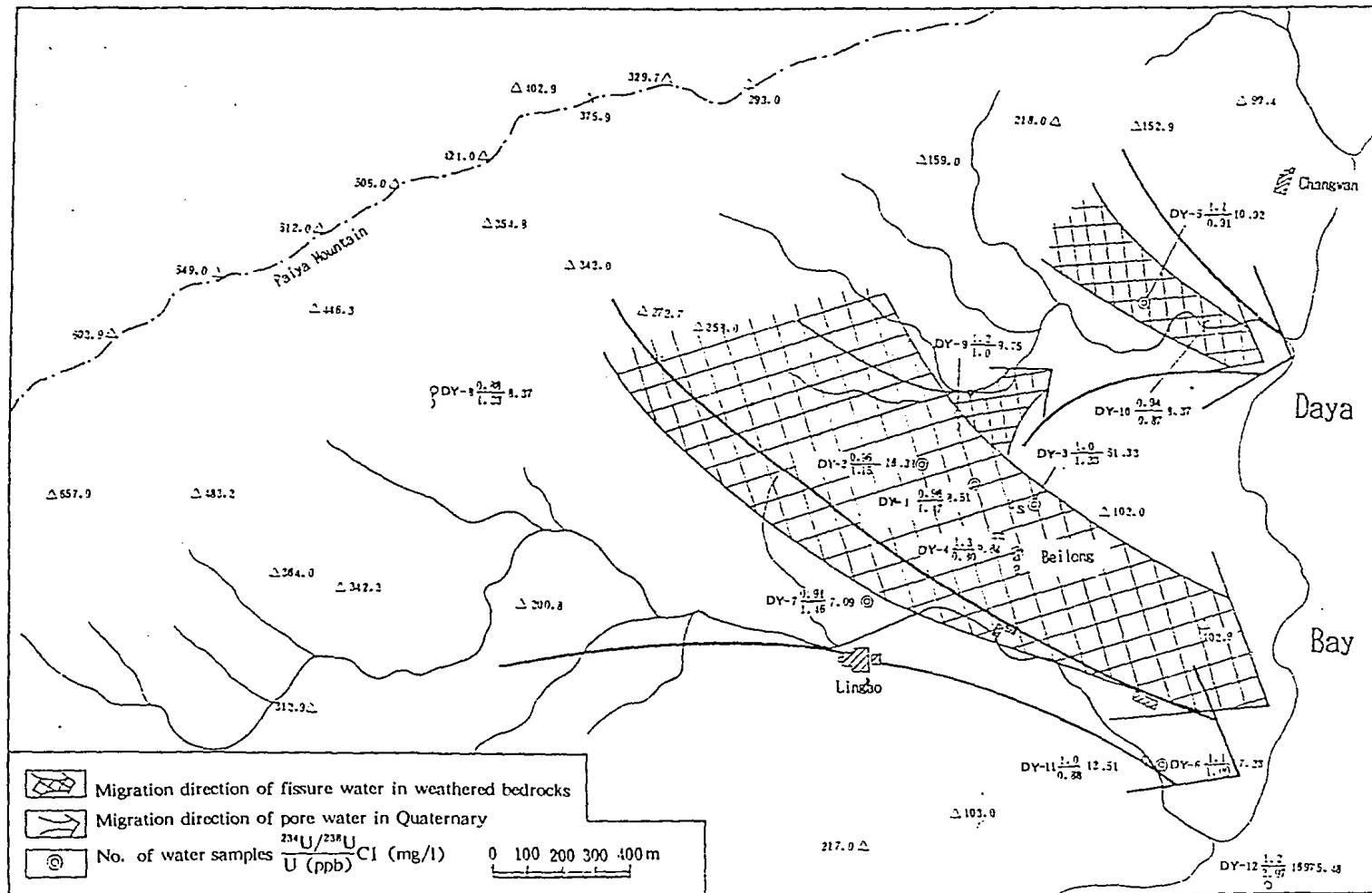


Fig. 3 Migration Direction of Groundwater in Ling'ao-Changwan Area

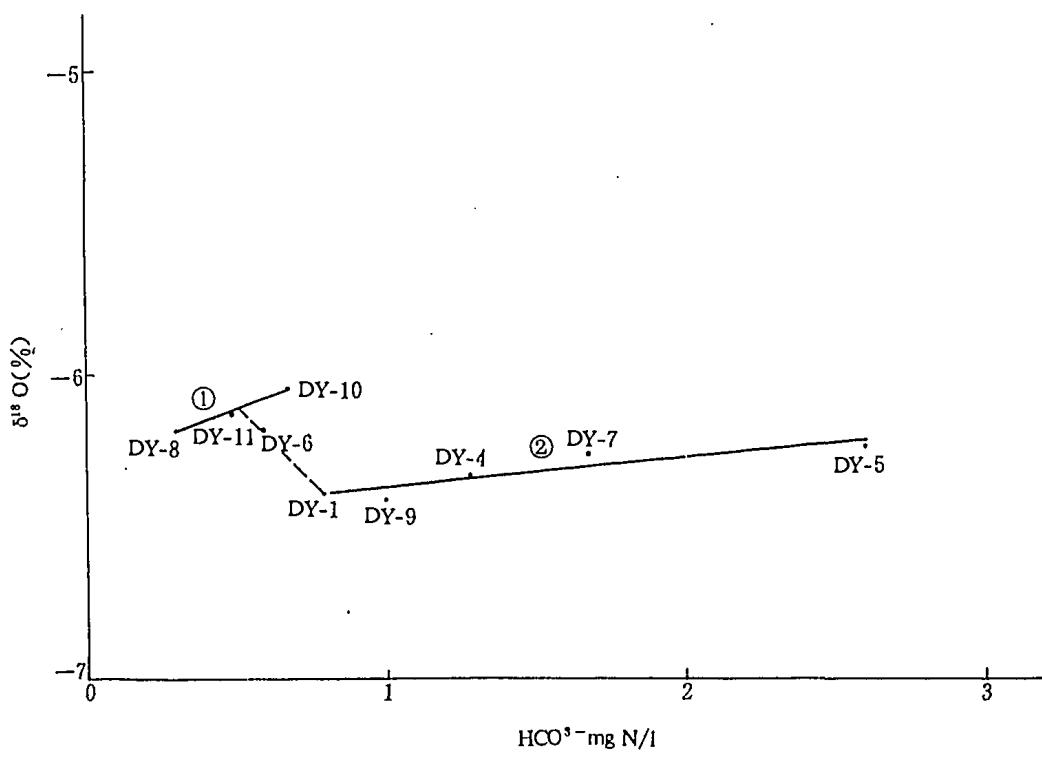


Fig. 4  $\delta^{18}\text{O}-\text{HCO}_3^-$  Relationship in Groundwater and Surface Water

## **PARTICIPANTS**

## French Participants

Thierry DUJARDIN	CEA/Siège
Amaury de BUZONNIERE	TA
Raymond CAPDEVIELLE	TA
Jean-Henri FORRESTIER	TA
Jean-Christophe GAUTHEY	TA
Gilles CHEVRIER	ANDRA
Jean-Claude FERNIQUE	ANDRA
Yves MARQUE	ANDRA
William BRAMY	SGN
Serge CARPENTIER	SGN
Patrice ROUX	SGN
Arsène SAAS	CEA/Cad

\* \* \*

Christian DESANDRE	TA
René VIENET	TA
Michel PELLOIE	FRA
Alfred MOLLER	Cogema/SGN
Pascale BON	Cogema/SGN
Bernard SOYER	Ambassade/France

## Chinese Participants

PAN Ziqiang	Bureau of Safety, Protection and Health, CNNC
LI Xuequn	Bureau of Safety, Protection and Health, CNNC
ZHU Zhiming	Bureau of Safety, Protection and Health, CNNC
QU Zhimin	Bureau of Safety, Protection and Health, CNNC
SUN Donghui	Beijing Institute of Nuclear Engineering, CNNC
SUN Mingsheng	Beijing Institute of Nuclear Engineering, CNNC
SONG Qianwu	Beijing Institute of Nuclear Engineering, CNNC
ZHOU Hanchen	Beijing Institute of Nuclear Engineering, CNNC
WEI Kuizi	Beijing Institute of Nuclear Engineering, CNNC
LI Jianhan	Beijing Institute of Nuclear Engineering, CNNC
WANG Wentao	Beijing Institute of Nuclear Engineering, CNNC
HE Chunying	Beijing Institute of Nuclear Engineering, CNNC
LI Tingjun	Beijing Institute of Nuclear Engineering, CNNC
ZHAO Yunhua	Beijing Institute of Nuclear Engineering, CNNC
YU Chengze	China Institute of Atomic Energy, CNNC
YAN Kezhi	China Institute of Atomic Energy, CNNC
WEI Jiang	China Institute of Atomic Energy, CNNC
QI Guangmao	China Institute of Atomic Energy, CNNC
XU Sikun	China Institute of Atomic Energy, CNNC
YUN Guichun	Institute of Nuclear Energy Technology, Tsinghua University
WU Jiaquan	Institute of Nuclear Energy Technology, Tsinghua University
GENG Zuohong	Institute of Nuclear Energy Technology, Tsinghua University
ZHUO Fengguan	Standardization Institute of Nuclear Industry, CNNC
GAO Mili	Standardization Institute of Nuclear Industry, CNNC
WANG Yongping	China Institute of Nuclear Industry Economics, CNNC
WU Chunxi	China Nuclear Information Center, CNNC
ZHAO Yamin	National Environmental Protection Administration
FAN Xuanlin	Beijing Huaqing Corporation, CNNC
XU Guoqing	Beijing Research Institute of Uranium Geology, CNNC
WANG Zhiming	Beijing Research Institute of Uranium Geology, CNNC
CHEN Zhangru	Beijing Research Institute of Uranium Geology, CNNC
REN Xianwen	China Institute for Radiation Protection, CNNC
HUANG Yawen	China Institute for Radiation Protection, CNNC
GU Cunli	China Institute for Radiation Protection, CNNC
GUO Liangtian	China Institute for Radiation Protection, CNNC
WANG Hui	China Institute for Radiation Protection, CNNC

CUI Anxi	China Institute for Radiation Protection, CNNC
TLAN Shuxing	China Institute of Nuclear Power, CNNC
YANG Shu	China Institute of Nuclear Power, CNNC
ZHANG Shilong	Lanzhou Nuclear Fuel Complex, CNNC
HU Kai	Lanzhou Nuclear Fuel Complex, CNNC
GUO Jiaping	Daya Bay Nuclear Power Plant, CNNC
ZHAO Yongsheng	Qinshan Nuclear Power Plant, CNNC
ZHU Peizhong	Qinshan Nuclear Power Plant, CNNC
CAO Li	Qinshan Nuclear Power Plant, CNNC

#### **Specially Invited Representatives**

LI Dingfan	CNNC
QIAN Gaoyun	Commission of Science and Technology, CNNC
WANG Chuanying	Commission of Science and Technology, CNNC
YAN Shuheng	Bureau of Science and Technology, CNNC
ZHENG Hualing	Bureau of Science and Technology, CNNC
ZHANG Zhifeng	Bureau of Nuclear Fuel, CNNC
REN Hailiang	Bureau of Nuclear Fuel, CNNC
LIN Sen	Bureau of Nuclear Fuel, CNNC
WANG Chengxiao	Bureau of Project and Planning, CNNC
QIU Xianfen	Dept. of Policy Study, CNNC
DONG Baotong	Dept. of Policy Study, CNNC
SHAN Guoxing	Bureau of Nuclear Power, CNNC
LUO Shirong	China Nuclear Energy Industry Corporation, CNNC
JIN Xiulan	China Nuclear Energy Industry Corporation, CNNC
YANG Yu	China Nuclear Information Center, CNNC
WANG Xiande	Beijing Institute of Nuclear Engineering, CNNC